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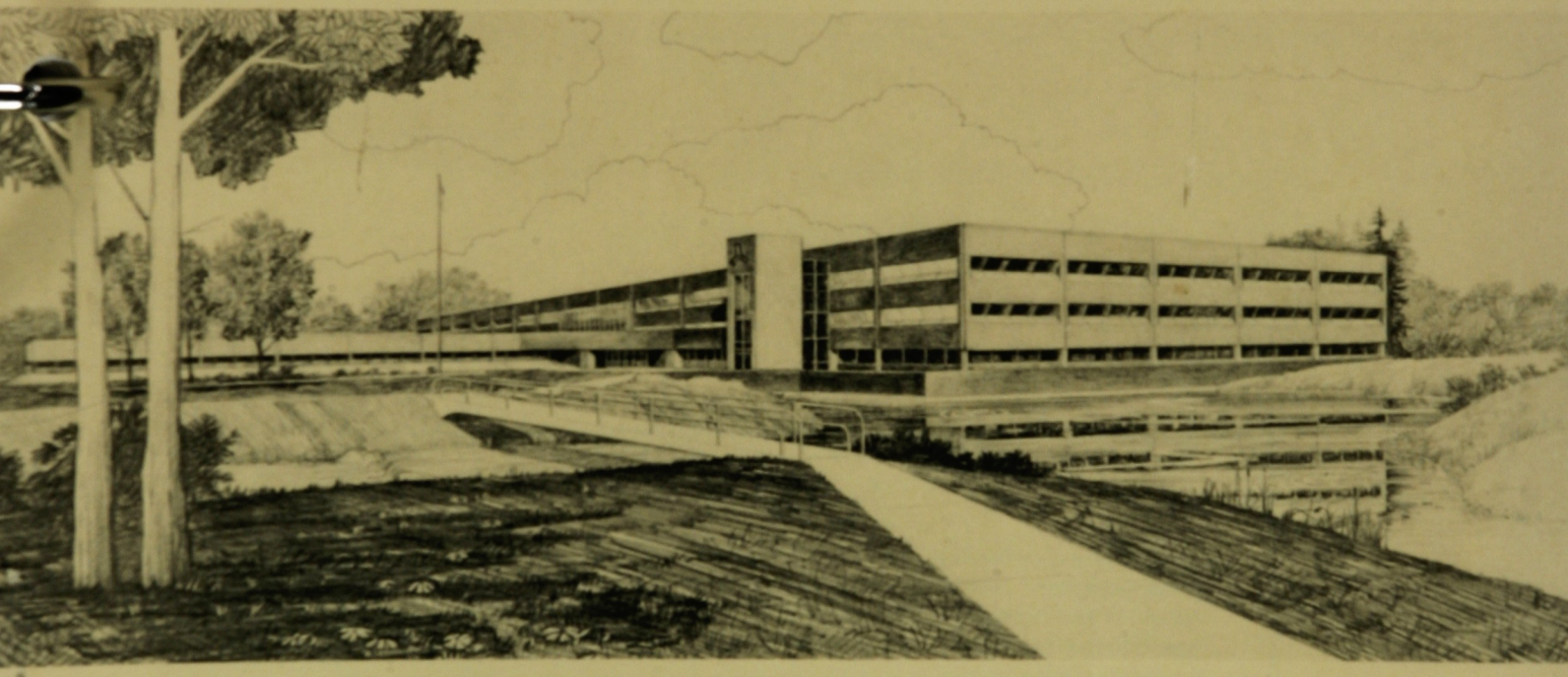
Idaho National Engineering Laboratory
Operated by the U.S. Department of Energy

Informal Report



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EG&G Idaho

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ABSTRACT

This report presents the chemistry and estimated behavior of tellurium during and after the accident at Three Mile Island Unit-2. The discussion of tellurium behavior is based on all available measurement data for ^{129m}Te, ^{132}Te, stable tellurium (^{126}Te, ^{128}Te, and ^{130}Te), and best estimate calculations of tellurium release and transport.

Results from Oak Ridge National Laboratory (ORNL) tests, Power Burst Facility (PBF) Severe Fuel Damage Tests at Idaho National Engineering Laboratory (INEL) and SASCHA tests from Karlsruhe, W. Germany are compared with calculated release fractions and samples taken from TMI Unit-2.

It is concluded that very little tellurium was released and transported from the TMI-2 core, probably as a result of holdup by zircaloy cladding and other structural materials.

SUMMARY

Thermodynamic calculations indicate that H_2Te is the predominant vapor species which results from the presence of excess hydrogen and high total pressure (8.2 to 15.2 MPa) in the upper plenum of TMI Unit-2. Increasing the system temperature will tend to dissociate H_2Te. However, temperatures $\geq$ 1200 K are needed for this to occur. The tellurium behavior presented in this report is based on all available measurement data for ^{129m}Te, ^{132}Te, stable tellurium (^{126}Te, ^{128}Te, and ^{130}Te), and best estimate calculations of tellurium release and transport. The predicted release was calculated using current techniques that relate release rate to fuel temperature and holdup of tellurium in zircaloy until significant oxidation occurs. The calculated release fraction was low (10%), but the total measured release for samples analyzed to date is about 5.8%. Of the measured tellurium in the containment sump water, upper plenum assembly surfaces, containment solids in the sump water, makeup and purification demineralizer, containment inside surface, and the reactor primary coolant there was about 2.4, 1.8, 0.88, 0.42, 0.17 and 0.86% of core inventory, respectively. A significant fraction (54%) of the tellurium predicted to be retained on the upper plenum surfaces (5.4% of the core inventory) was deposited during the high pressure injection of coolant at about 200 min after the reactor scram. Comparison of tellurium behavior with in-pile and out-of-pile tests suggests that zircaloy holds tellurium until significant cladding oxidation occurs. Analyses of samples from the core region of TMI-2 indicate that about 49% of core inventory is retained in the surface of the debris bed. Core samples taken from 0.28 to 0.94 m into the debris bed contained lower amounts of tellurium, suggesting that a highly volatile tellurium species was released from the hot debris bed and deposited in the cooler surface debris bed. No correlation was found between the atoms of tellurium and those of tin, zirconium, iron, chromium, and nickel.

ACKNOWLEDGMENTS

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ACRONYMS AND ABBREVIATIONS

FSAR	Final Safety Analysis Report
GPU	General Public Utilities
ICP	Inductively coupled plasma
ICP-AES	Inductively coupled plasma excitation source
INEL	Idaho National Engineering Laboratory
LWR	Light water reactor
ORNL	Oak Ridge National Laboratory
PBF	Power Burst Facility
PWR	Pressurized water reactor
SFD	Severe fuel damage
SFD-ST	Severe fuel damage scoping test
TGT	Thermal gradient tube

TELLURIUM CHEMISTRY, TELLURIUM RELEASE AND DEPOSITION DURING THE TMI-2 ACCIDENT

INTRODUCTION

Until recently, studies of tellurium release from the reactor core were based on temperature, and its volatility in comparison with other potentially important radionuclides (I, and Cs).[1] However, tests at the core melt facility at Oak Ridge National Laboratory (ORNL), the Severe Fuel Damage (SFD) Tests at the Idaho National Engineering Laboratory (INEL), and the SASCHA Tests at Karlsruhe suggest that tellurium may be held up by zircaloy cladding and result in significantly lower release from the core. These tests also demonstrated that tellurium releases increase significantly when the zircaloy cladding is oxidized and the previously held up tellurium is released. Lorenz et al.,[2] emphasized that a lower-than-expected tellurium release from the core does not necessarily mean a lower calculated release to the environment, but rather that the tellurium transport pathway is different from that previously envisioned, and higher or lower releases to the environment may result depending on zircaloy oxidation during an accident progression. Also, Elrick and Sallach[3] indicate that tellurium may react with structural materials (stainless steel) and thereby be retained in the primary coolant system. Silver (from the control rods) and tin (a minor constituent of zircaloy cladding) aerosols are efficient scavengers of tellurium vapor.[3]

The tellurium behavior during and after the Three Mile Island-Unit 2 accident may shed further light on tellurium transport during a severe accident. The accident at TMI-2 on March 28, 1979, resulted in severe damage to the reactor core. As a consequence, numerous data are being gathered to study fission product behavior during and after the accident. A large data base with supporting analyses exists for noble gases, iodine, and cesium behavior. Most of the radiological and chemical analyses of the samples from TMI-2 (especially those taken soon after the accident) sparingly report tellurium levels. This lack of data is probably due to the fact that special analytical methods are required to determine the low tellurium concentrations in highly radioactive samples; unless a special

effort is made, the tellurium content is usually not measured. Atomic emission spectroscopy using an inductively coupled plasma excitation source (ICP-AES) was used at INEL to analyze tellurium in TMI-2 leadscrew and core debris samples. A limited number of samples drawn from primary coolant water, reactor coolant bleed tank water, the containment inside surface, the containment sump water and solids, containment atmosphere, auxiliary building sump water, makeup and purification system, core debris bed and upper plenum assembly surfaces were analyzed for tellurium.

The objectives of this report are to present the results of tellurium analyses performed to date on TMI-2 samples, estimate the tellurium distribution and the release and deposition fractions, and compare the data with current best estimate behavior models and data from out-of-reactor and in-reactor tests.

The following sections present:

- Calculations of tellurium chemistry,
- Measurements of tellurium release from the TMI-2 Core,
- Tellurium release data from ORNL tests,
- Tellurium releases from the PBF Severe Fuel Damage tests and the SASCHA tests,
- Comparisons of the TMI-2 tellurium behavior with the above out-of-pile and in-pile tests,
- Conclusions reached in this investigation.

Appendicies A and B present tellurium analytical methods and TRAP-MELT input deck, respectively.

TELLURIUM CHEMISTRY

Tellurium volatility in the primary coolant system of a nuclear reactor is greatly affected by the environment (oxidizing or reducing), type of chemical species and interaction of these species with other core materials and fission products. In neutral or reducing conditions, the most stable tellurium species are Te, Te_2 and H_2Te. Under oxidizing conditions, oxidized Species TeO, TeO_2, and $TeO(OH)_2$ become dominant.

The equilibrium concentrations of these vapor species except $TeO(OH)_2$ were calculated[4] using the FLUEQU code developed at Sandia National Laboratory.[5] The input data shown in Figure 1 are the hydrogen-to-steam and tellurium-to-oxygen ratios for various times during the TMI-2 accident. In Figure 1, the square roots of tellurium-to-oxygen ratios were used to facilitate presentation of the data. These ratios were calculated from the estimated hydrogen and tellurium releases and steam flow rates during the TMI-2 accident. The correlation of the tellurium release with an excess of hydrogen is clearly evident.

Two temperatures and two pressures were considered. The lower temperature 755 K (900°F) is considered to be a representative temperature for the upper plenum assembly and the higher temperature 1255 K (1800°F) is typical of the region at the top of the core. The higher pressure was 15.2 MPa (2250 psi) which represents the operating pressure and the lower pressure 8.2 MPa (1200 psi) is chosen to represent the conditions during the steam blowdown.

The calculated partial pressures of some tellurium chemical species at 8.2 MPa (1200 psi) and for the temperatures 755 (900) and 1255 K (1800°F) are shown in Figures 2 and 3, respectively. The preponderant tellurium species is H_2Te, except when there is little hydrogen present during the burst of steam occurring at 173-178 min. Thus the chemistry of tellurium under TMI-2 accident conditions should be determined by the reactions of H_2Te vapor with the materials of the reactor system.

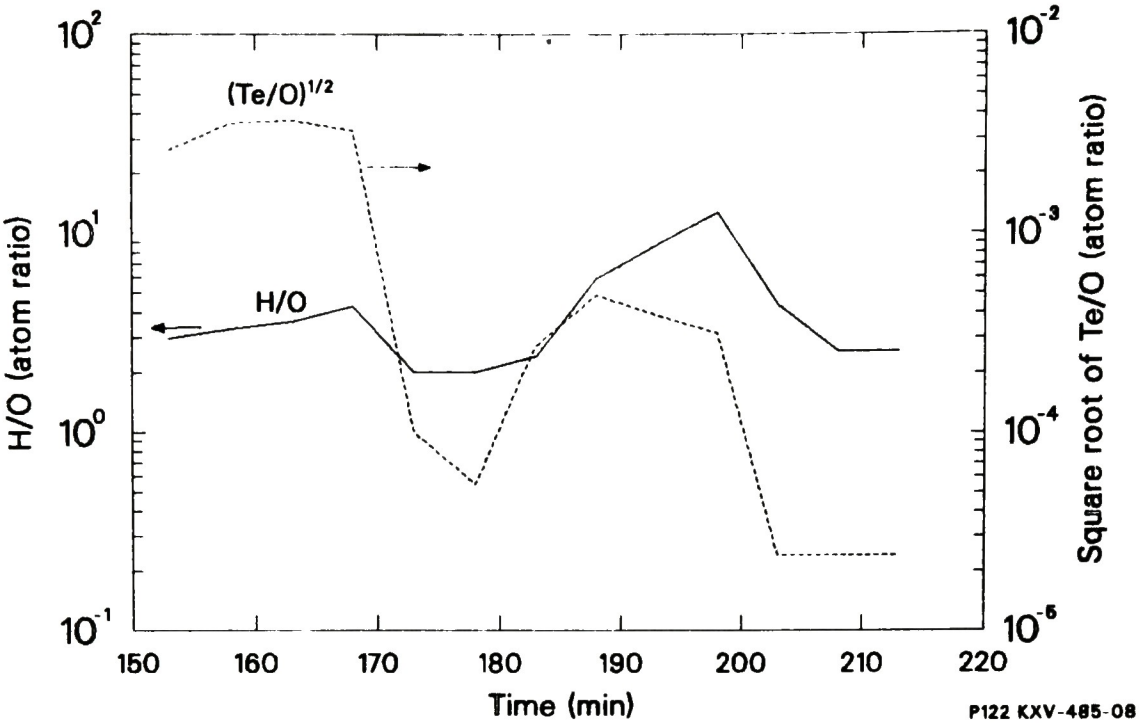

Figure 1. The (H/O) and $(Te/O)^{1/2}$ versus time.

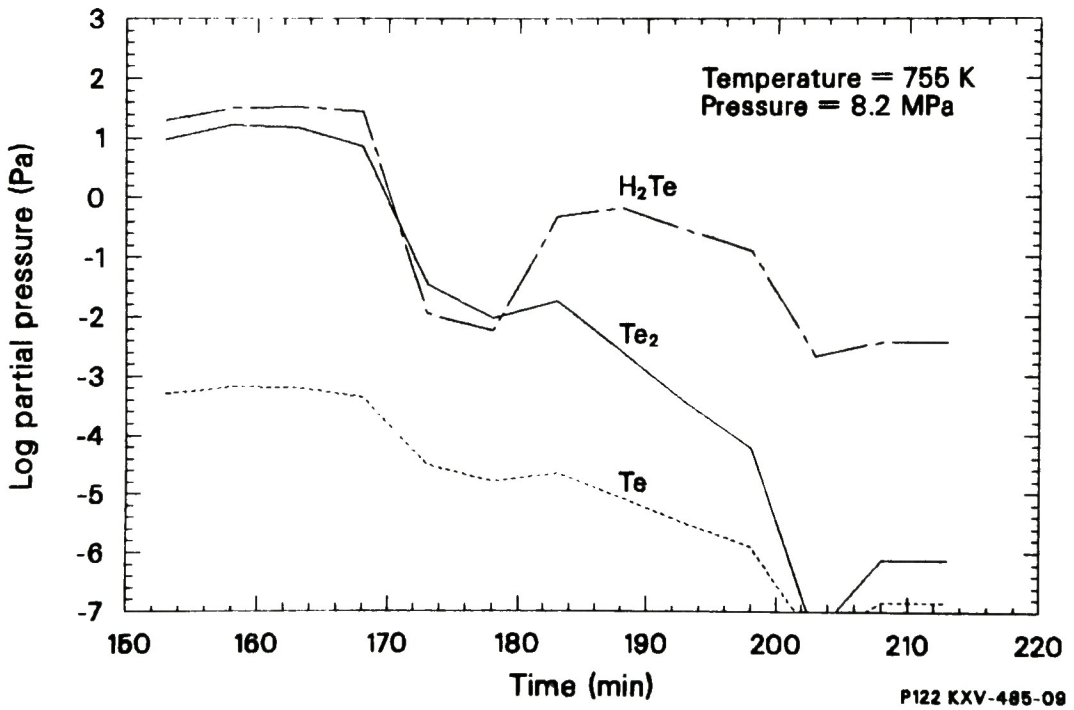

Figure 2. Partial pressures of tellurium species at 755 K and 8.2 MPa total pressure.

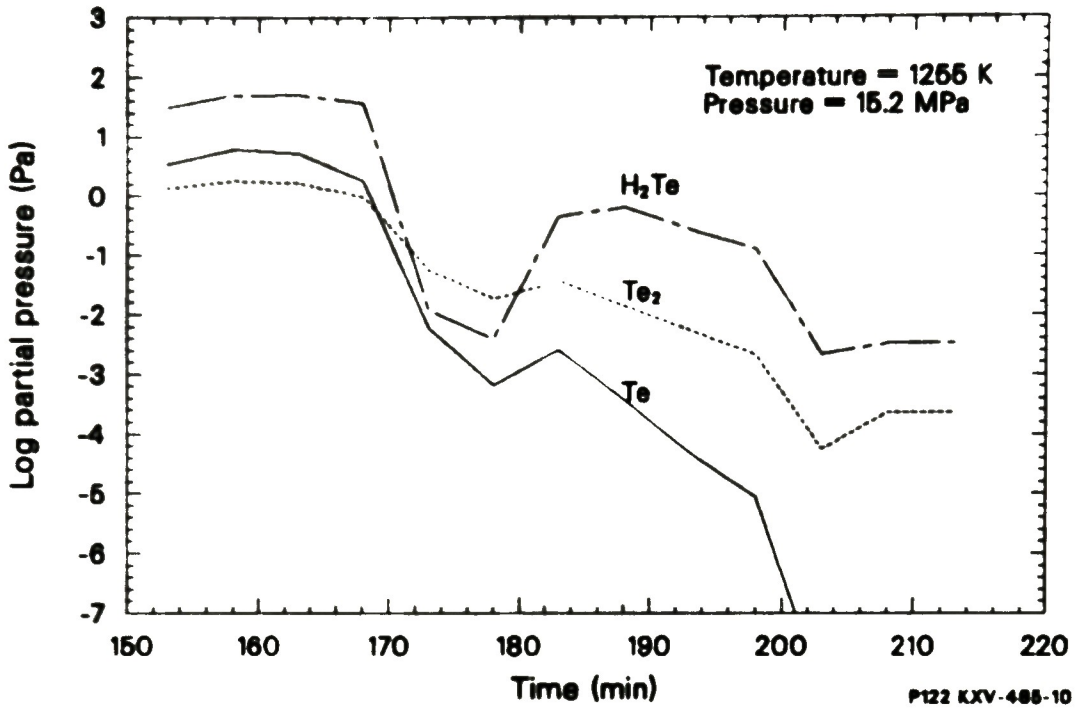

Figure 3. Partial pressures of tellurium species at 1255 K and 15.2 MPa total pressure.

Several calculations were made with a broader range of parameters for general trends. Selected data are presented in Figures 4 through 9. As shown in Figures 4 and 5, the vapor composition in steam, with no excess hydrogen at atmospheric pressure (0.1 MPa), consists of Te_2 at a temperature of 800 K and Te at a temperature of 1200 K. These elemental vapors persist as the dominant species until about 20% of the steam has reacted (H/O = 2.5) when the total pressure is equal to the atmospheric pressure (0.1 MPa). As the total pressure is increased, the Species H_2Te is formed at the expense of elemental vapor species as shown in Figures 6 and 7.

The effect of temperature for an H/O ratio and pressure where H_2Te is the predominant species at 800 K is shown in Figure 8. As the temperature increased to 2000 K, thermal dissociation of H_2Te takes place and the elemental forms again become dominant.

The effect of dilution of the tellurium content is shown in Figure 9. The Species Te_2 becomes unstable with respect to Te. However, the H_2Te species is also seen to become the major species even for steam/hydrogen mixtures in which the H/O ratio is as low as 2.04.

In summary, the formation of the vapor Species H_2Te is favored by the presence of excess hydrogen and high total system pressures. This species is also favored when the tellurium content of the gas phase is diluted (reduced). Increasing the system temperature will tend to dissociate H_2Te but temperatures $\geq$1200 K are needed depending on other system parameters.

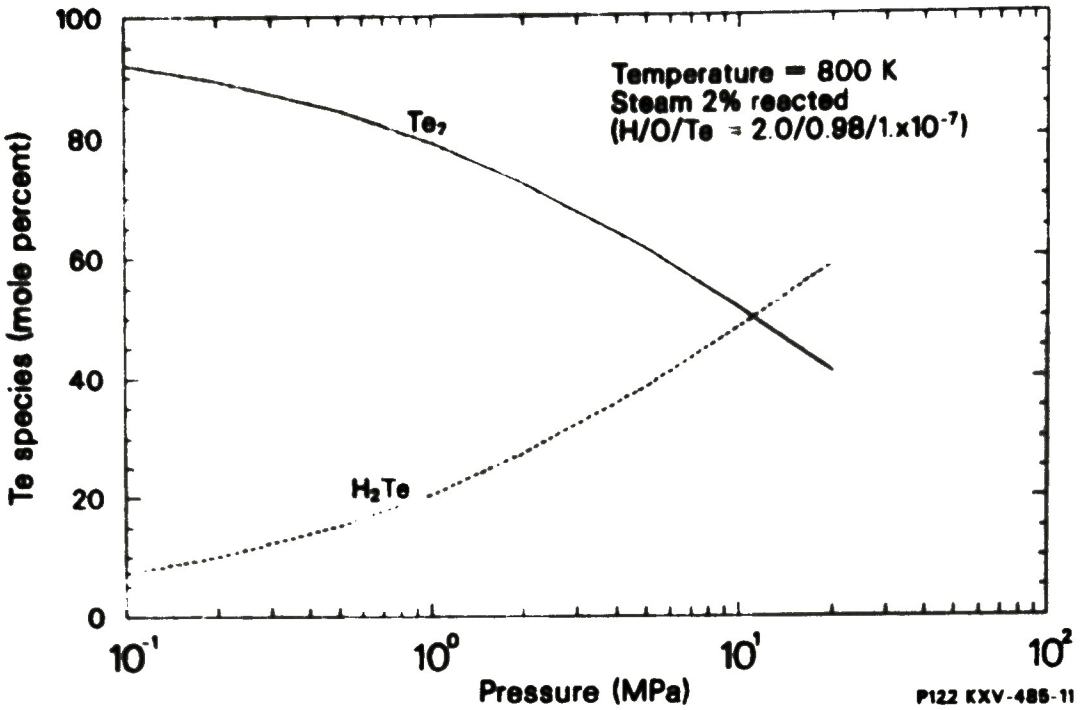

Figure 4. Effect of total pressure and composition of tellurium at 800 K and with (H/O) = 2.04.

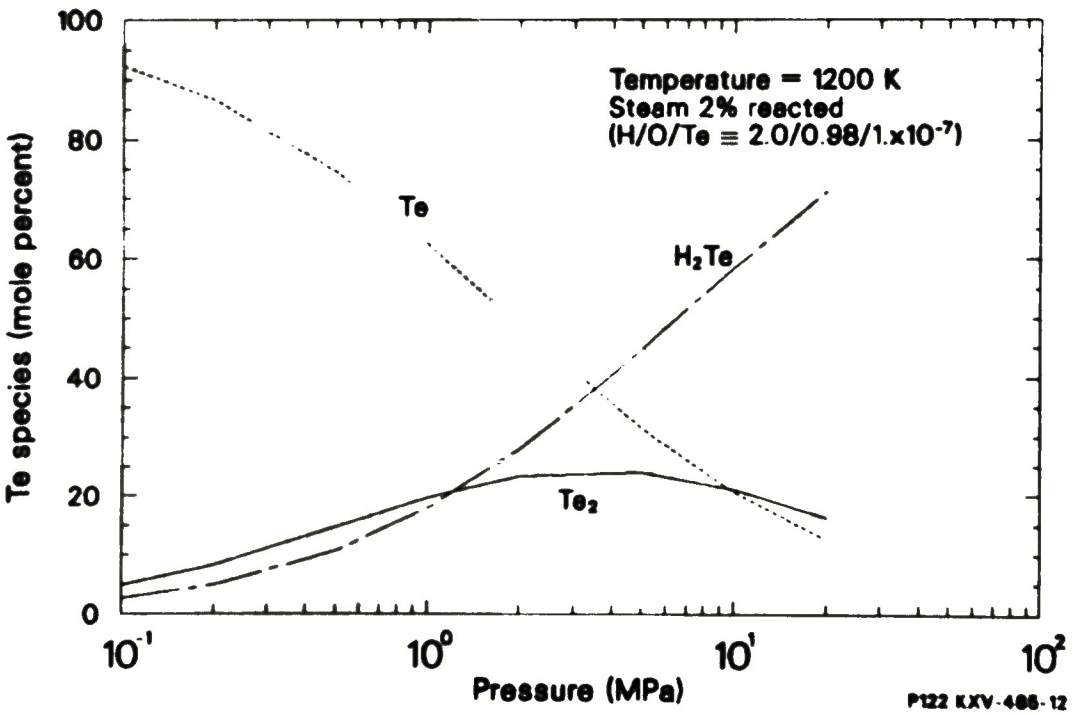

Figure 5. Effect of total pressure on composition of tellurium species at 1200 K and with (H/O)= 2.04.

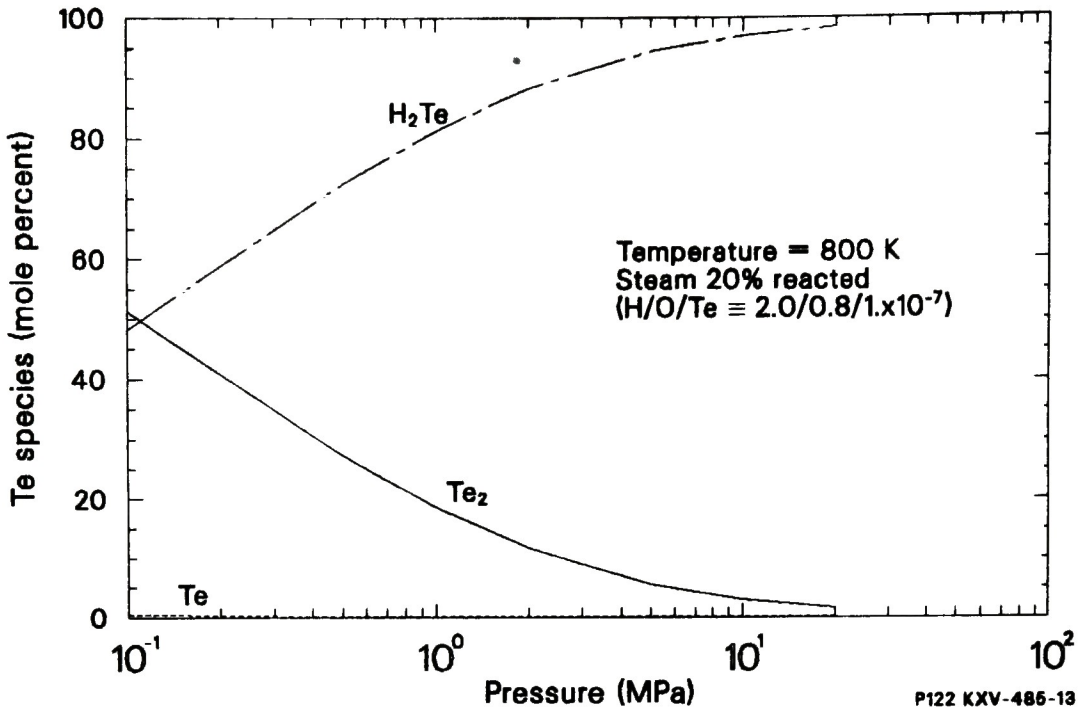

Figure 6. Effect of total pressure on composition of tellurium species at 800 K and with (H/O)= 2.5.

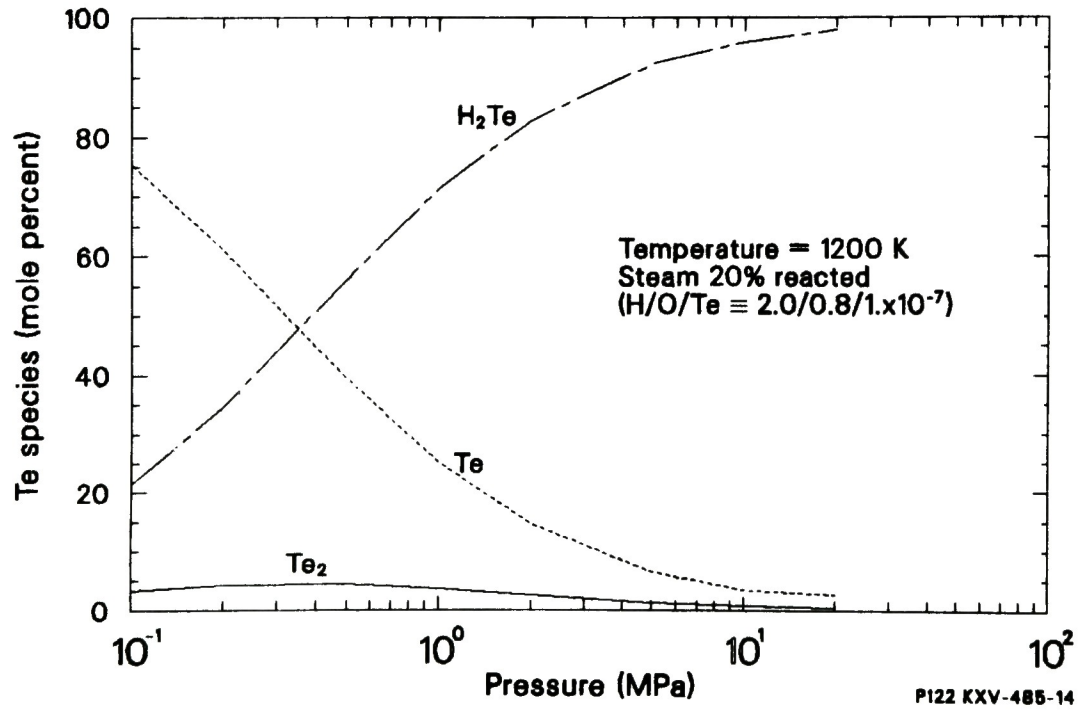

Figure 7. Effect of total pressure on composition of tellurium species at 1200 K and with (H/O)= 2.5.

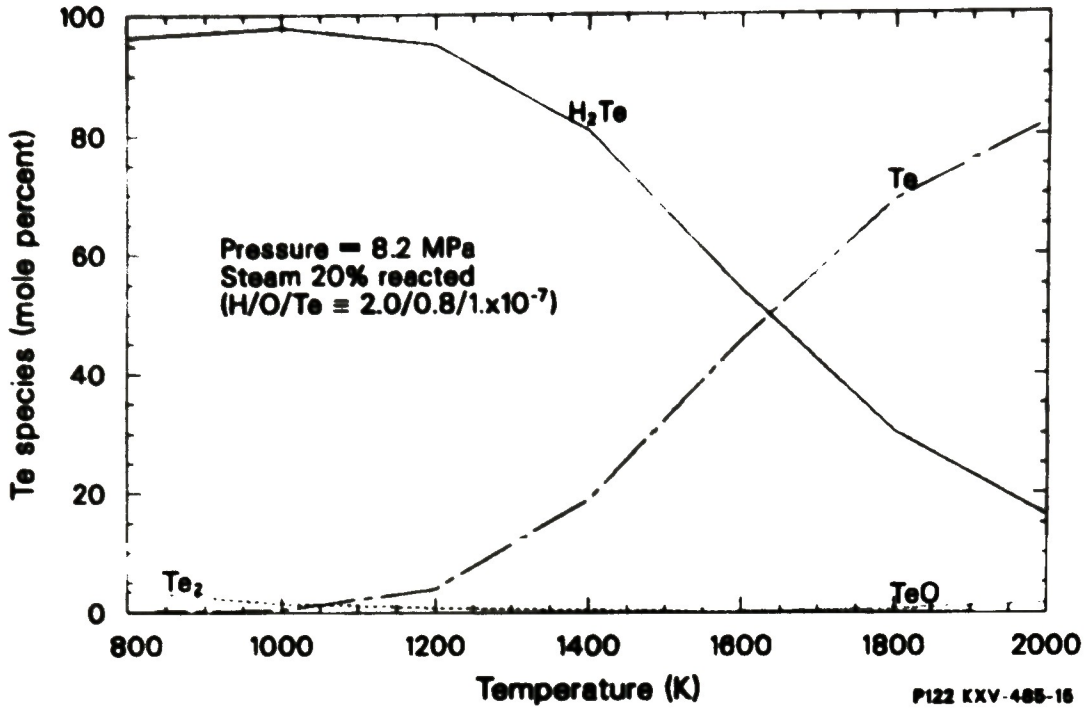

Figure 8. Effect of temperature on tellurium species.

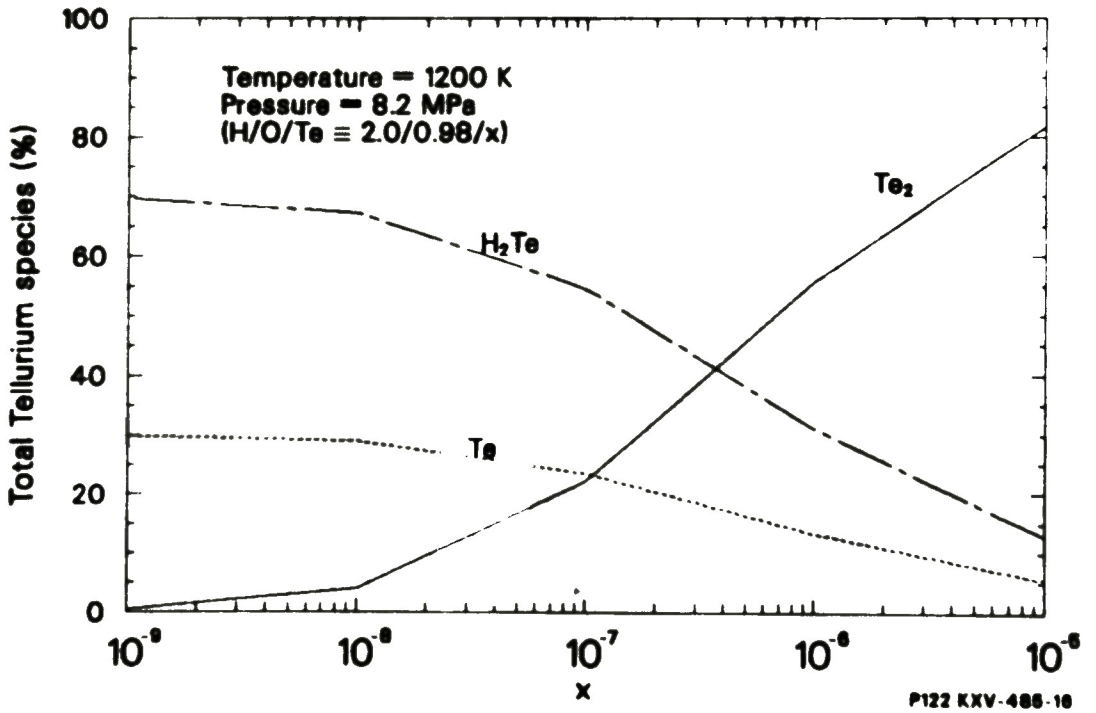

Figure 9. Effect of dilution on composition of tellurium species.

TELLURIUM RELEASES

Measurements of Tellurium Released from TMI-2 Core

A summary of tellurium measured in samples taken to date from the TMI-2 plant systems and components is listed in Tables 1, 2, and 3. The systems and components included in the tellurium investigation were:

- Reactor primary coolant,[6,7]
- Reactor coolant bleed tank water,[8]
- Containment inside surface,[7,9]
- Containment sump water and solid debris,[10-13]
- Containment atmosphere,[14]
- Auxiliary building sump water,[15]
- Makeup and purification filter,[16,17]
- Upper plenum surfaces[18] (samples from the H8 and B8 leadscrews), and
- Core debris (grab samples).

Approximately 0.086% of the ^{132}Te core inventory was determined to be in the primary coolant water, based on an early analysis (March 29, 1979) of water samples.[6] This low tellurium inventory may be due to: (a) low tellurium release, (b) low tellurium solubility in water, or (c) the retention of tellurium in the core.

About 9×10^{-3}% of the core inventory was estimated to be in the reactor coolant bleed tank, based on the data obtained on December 8, 1979, by ORNL.[8]

TABLE 1. SUMMARY OF TELLURIUM RELEASE FRACTIONS IN TMI-2 SYSTEMS

System or Component	Sampling Data	Tellurium Isotope	Percent of Initial Core Inventory[a]	Reference
Reactor primary coolant	3-29-79	^{132}Te	0.086	(6)
	3-30-79	^{132}Te	0.086	(6)
	4-10-79	^{132}Te	0.010	(6)
	6-21-79	^{132}Te	0.014	(7)
Reactor coolant bleed tank water	12-18-79	^{129}Te	0.009	(8)
Containment inside surface	8-29-79	^{127}Te	0.045	(9)
	8-29-79	^{129m}Te	0.12	(7)
	9-09-79	^{129m}Te	0.17	(9)
Containment sump water	6-20-79	^{129}Te	1.06	(10)
	6-20-79	^{132}Te	2.40	(10)
	8-29-79	^{129m}Te	0.008	(11)
Solids in water	8-28-79	^{129m}Te	0.47	(11)
			0.88	(12)
Sludge[b]	10-26-82	^{130}Te	765 ppm	(13)
		^{128}Te	108 ppm	(13)
		^{126}Te	27 ppm	(13)
Containment atmosphere	5-1-80	^{129m}Te	3.5×10^{-6}	(14)
Auxiliary building sump tank water sample	3-25-80	^{127m}Te	1.3×10^{-4}	(15)
		^{129m}Te	1.5×10^{-3}	(15)
Makeup and purification demineralizer	May 1983	Stable Te	4.2×10^{-1}	(16,17)
	May 1983	Stable Te	3.1×10^{-1}	(16,17)
Upper plenum surface	September 1984	Stable Te, Te inpurities in stainless steel[d]		
H8 leadscrew			1.2, 1.8	(18)
B8 leadscrew			0.52	(18)

TABLE 1. (continued)

System or Component	Sampling Data	Tellurium Isotope	Percent of Initial Core Inventory[a]	Reference
Surface of the core debris bed	Batch 1 (Samples 1-6)[c] Sept-Oct/83 Batch 2 (Samples 7-11)[c] Mar-Apr/84	Stable Te, Te impurities in stainless steel[d]	48.8[c]	--
Total release from the reactor core[e]			5.8	

a. Core inventory calculated by ORIGEN-2 code (Reference 19) and decayed to time of sample analysis.

b. Not analyzed because the data were semiquantitative.

c. See Table 3.

d. Tellurium impurity in the stainless steel components in the core was estimated to be about 804 g and the calculated stable tellurium inventory was about 3649 g.

e. The sum of largest tellurium releases from the reactor core measured in the plant system.

TABLE 2. CORE DEBRIS BED SAMPLES AND ELEMENTAL CONCENTRATIONS

Sample Number	Subsample Number	Subsample Weight (mg)	Description (μm particles)	Concentration (mg/g) Te	Sn	Zr	U
1[a]	1B	15	>4000	7.6	--[b]	12.9	830.7
1	1E	29	>4000	5.4	10.0	655.5	177.1
1	1F	20	1680-4000	5.5	--[b]	136.8	570.0
1	1H	33	1680-4000	4.3	--[b]	367.4	472.5
1	1	13	74-149	8.8	9.1	258.5	329.0
3[c]	3B	10	>4000	4.3	--[b]	257.5	<13[b]
3	3	12	30-74	9.5	--[b]	355.0	317.0
4[d]	4A	22	>4000	6.5	11.4	134.1	938.6
4	4C	38	>4000	3.8	--[b]	12.3	514.5
4	4E	15	>4000	9.5	--[b]	26.1	883.4
5[e]	5C	28	>4000	4.1	7.1	39.4	574.0
5	5G	75	1680-4000	0.9	--[b]	18.9	508.8
5	5H	42	1000-1680	2.7	4.8	168.1	343.0
5	5I	36	1000-1680	5.4	5.9	2.4	440.0
6[f]	6J	77	1000-1680	2.0	--[b]	538.7	128.3
8[g]	8	10	20-30	13.7	--[b]	129.6	487.2
9[h]	9B	33	>4000	3.5	6.1	220.0	504.2
7,10,11[i]	--	--	--	--	--	--	--

a. Out of 16 subsamples from Sample 1, only 5 subsamples contained measurable tellurium.

b. Below the detection limit.

c. Out of 18 subsamples from Sample 3, 2 subsamples contained measurable tellurium.

d. Out of 5 subsamples from Sample 4, 3 subsamples contained measurable tellurium.

e. Out of 8 subsamples from Sample 5, 4 subsamples contained measurable tellurium.

f. Out of 15 subsamples from Sample 6, only 1 subsample contained measurable tellurium.

g. Out of 18 subsamples from Sample 8, only 1 subsample contained measurable tellurium.

h. Out of 13 subsamples from Sample 9, only 1 subsample contained measurable tellurium.

i. None of 18 subsamples from Samples 7 and 11, and 24 subsamples from Sample 10 contained measurable tellurium.

TABLE 3. RETAINED TELLURIUM FRACTION IN THE CORE DEBRIS BED

Sample	TMI-2 Core Location	Location of the Sample in Debris bed	Sample Weight (g)	Thickness of Debris Bed Layer h in m	Fraction of Subsamples That Contained Tellurium in Percent[a] (ratio)	Average Concentration in gram per gram	Retained Tellurium Fraction in Percent[b]
1	H8	Surface	70.88	0.027	31.0 (5/16)	2.0×10^{-3}	39.0
2	H8	Near Surface, 0.076 m (3-in.)	126.20	--	--[c]		--[c]
3	H8	0.559 (22-in) into the debris bed	152.71	0.035	11.0 (2/18)	7.7×10^{-4}	19.4
4	E9	Surface	16.59	0.017	60.0 (3/5)	4.0×10^{-3}	48.8
5	E9	Near Surface, 0.076 m (3-in.)	90.96	0.029	40. (4/10)	1.3×10^{-3}	27.3
6	E9	0.559m (22-in.) into the debris bed	140.7	0.034	7.0 (1/15)	1.3×10^{-4}	3.3
7	H8	0.356m (14 in.) into the debris bed	135.86	--	0.0 (0/18)	0.0	0.0
8	H8	0.669m (27.5 in.) into the debris bed	152.76	0.035	6.0 (1/18)	7.6×10^{-4}	19.1
9	H8	0.775m (30.5 in.) into the debris bed	153.0	0.035	8.0 (1/13)	2.7×10^{-4}	6.8

TABLE 3. (continued)

Sample	TMI-2 Core Location	Location of the Sample in Debris bed	Sample Weight (g)	Thickness of Debris Bed Layer h in m	Fraction of Subsamples That Contained Tellurium in Percent[a] (ratio)	Average Concentration in gram per gram	Retained Tellurium Fraction in Percent[b]
10	E9	0.737m (29.0 in.) into the debris bed	173.90	--	0.0 (0/24)	0	0.0
11	E9	0.940m (37.0 in.) into the debris bed	148.75	--	0.0 (0/18)	0	0.0

a. Number of subsamples containing tellurium per 100 subsamples analyzed.

b. Fraction of core inventory in percent. Uncertainty = ±60%.

c. Analyzed by B&W, and no tellurium analysis was performed.

Plug "401"[a] was gamma scanned at ORNL on August 29, 1979. It is assumed that the disk was representative of the entire inner containment surface area of 2.2×10^8 cm^2. Analysis[7] of this indicated that ~0.045 and 0.12% of ^{127m}Te and ^{129m}Te core inventories, respectively, were deposited on inside surfaces of the containment building. Tellurium plateout on the hydrogen recombiner inlet spool piece was analyzed.[9] The hydrogen recombiner assembly located outside the containment is connected to the containment by piping. The inlet spool piece was analyzed for ^{129m}Te and the results indicated that 0.17% of the ^{129m}Te core inventory was deposited on containment surfaces. The effective area of the containment was estimated in a very approximate manner; the release fraction is probably no better than an order-of-magnitude estimate.

The containment sump water was analyzed[10] on June 20, 1979. The estimated inventory of ^{129m}Te and ^{132}Te were 2.4 and 1.69×10^{-5} μCi/mL. Based on the containment sump volume of 2.16×10^9 mL and the core inventory on June 20, 1979 (4.9×10^{11} μCi for ^{129m}Te, and 1.53×10^6 μCi for ^{132}Te), the estimated release fractions of ^{129m}Te and ^{132}Te are 1.06 and 2.4%, respectively.

Water samples (30 mL) from the top, middle, and bottom of the containment sump were analyzed on August 28, 1979, at ORNL.[11] About 0.035 μCi/mL of ^{129}Te was detected in the bottom sample. Two samples of solid debris collected with the bottom water sample were taken; they were centrifuged, washed, and gamma scanned. About 0.277 and 0.514 μCi/mL of ^{129m}Te was detected. Based on the containment sump water volume (2.16×10^9 mL) and the tellurium core inventory (9.65×10^{11} μCi for ^{129}Te and 1.27×10^{11} μCi for ^{129m}Te),[12] on August 28, 1979, the estimated core release fractions of the water sample and two solid samples were 0.008, 0.47, and 0.88%, respectively. Semiquantitative spark source mass spectrometry was performed on the containment sludge sample on October 26, 1982, and identified 900 ppm (μg/g) of stable tellurium (^{126}Te, ^{128}Te, and ^{130}Te).[13]

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a. A painted steel disk of 7 cm (2.75 in.) diameter projecting into the containment building.

A containment building air sample, including associated suspended matter was analyzed during April 29 to May 2, 1980.[14] The air contained 5×10^{10} μCi/mL of ^{129m}Te. Based on the containment volume of 5.6×10^{10} mL, the estimated containment building inventory was 28 μCi, which represents 3.5×10^{-6}% of the core inventory of ^{129m}Te projected at this time by ORIGEN code.[19]

A water sample from auxiliary building sump Tank 2B was analyzed by ORNL on March 25, 1980.[15] About 1×10^{-3} μCi/mL of $^{\beta}Te$, beta emitting tellurium (which is a mixture of long lived ^{127m}Te and ^{129m}Te) was identified. This analysis was accomplished by chemical separation and subsequent beta counting of the sample which contained $^{\beta}Te$. Based on the auxiliary building sump Tank 2B volume (5.11×10^{7} mL) and the projected core inventory of nuclides on March 25, 1980, the estimated release fractions of ^{127m}Te and ^{129m}Te are 1.3×10^{-4} and 1.5×10^{-3}%, respectively. These release fractions are estimated assuming that the mixture of $^{\beta}Te$ contained 100% of either ^{127m}Te or ^{129m}Te.

The contents of the makeup and purification demineralizer were analyzed in May 1983.[16] General Public Utilities (GPU) has estimated[17] that 1.74×10^{8} (mL) (46,000 gal) of highly contaminated reactor coolant passed through the demineralizer vessels during the accident. The amounts of stable Te detected in the A and B demineralizers were 10 and 30 ppm, respectively. Based on the calculated inventory of stable tellurium of 3643 g and estimated masses of solid materials in A and B demineralizers (5.1×10^{5} g), the estimated release fractions are 0.14 and 0.42%, respectively.

Control rod mechanism leadscrew samples from the H8 and B8 positions were analyzed by an induction coupled plasma technique for elemental tellurium at INEL.[18] The radioactive tellurium nuclides are expected to have decayed to a negligibly small amount; measured tellurium would be the stable tellurium nuclides (^{126}Te, ^{128}Te, and ^{130}Te) from the fission products, and doped tellurium (tellurium added to stainless steels as a free-machining agent). The precise quantity of doped tellurium is

generally proprietary information; however, tellurium weight percentages of 0.0005 to 0.1% are typical. The estimated core inventory fraction retained on upper plenum assembly surfaces was 0.018.[18] The largest tellurium releases measured in the above plant systems and components were summed to give a total fractional release of about 5.8% of the core inventory.

Six samples of particulate debris were removed from the TMI-2 core debris bed during September-October 1983.[20] Subsequently in 1984, five more samples were obtained at the H8 (mid-core) and E-9 (mid radius) locations of the core as shown in Figure 10. The samples are from seven depths: surface of the debris bed (0 to 0.076 m), 0.356 m, 0.559 m, 0.699 m, 0.737 m, 0.775 m and 0.940 m deep in the bed. Ten samples were analyzed by EG&G and the remaining sample (sample 2 from 0.076 m deep at the H8 position) was analyzed by the Babcock and Wilcox research center.[21] A known mass of particles in each sample was dissolved in a strong acid and analyzed for tellurium by inductively coupled plasma (ICP) spectroscopy. ICP spectroscopy quantifies elemental rather than isotopic tellurium content, therefore this analysis method does not distinguish between tellurium from material sources (impurity in stainless steel and fission product tellurium). Although an estimated source of natural tellurium (804 g) is included in the core inventory, the nature of tellurium (natural or isotopic) measured in the core debris samples is unknown.

About 155 subsamples from ten debris samples were analyzed and the data are presented in Table 2. Only 18 subsamples contained tellurium above the detection limit. The tellurium detection limits are very high because of the dilution required to reduce the sample activity. The error in tellurium measurement was estimated to be about $\pm$60%.

The amount of tellurium retained in the core debris bed was estimated from

$$R(Te) = \frac{V\rho C}{I} \times 100 \qquad (1)$$

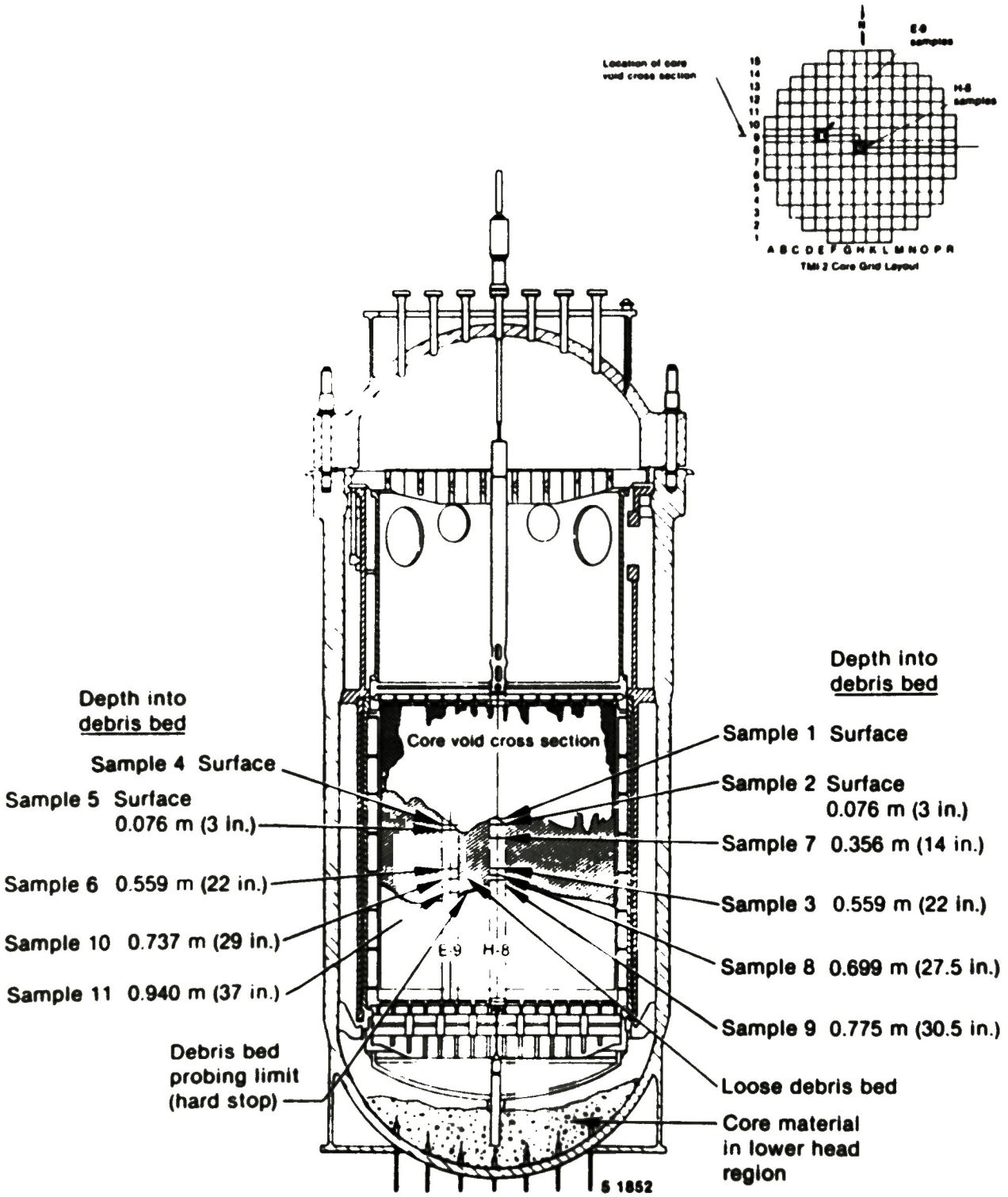

Figure 10. TMI-2 core debris grab samples.

where

$R(Te)$ = retained tellurium in percent

V = volume of debris bed layer in m^3

= $\pi(1.67)^2 h$ in m^3

h = thickness of debris bed layer from which a sample collected

= $(\text{sample weight}/\rho)^{1/3}$ in m,

ρ = debris bed density = 3.65×10^6 g/m^3

C = average concentration of tellurium in grams per gram of sample

I = tellurium inventory = 4453 g.

These results consisting of the thickness of debris bed layer, fraction of subsamples that contained tellurium, average concentration at each location in the debris bed, and the retained tellurium fraction are presented in Table 3. The maximum concentration of tellurium retained in the surface layer of the core was measured to be 4.0×10^{-3} g per gram of the sample. This concentration of retained tellurium was extrapolated to debris contents of the entire surface layer of about 0.029 m (1.15 in.) thick (9.27×10^5 g) and normalized to the calculated core inventory of tellurium (4453 g). The measured tellurium retained in the core debris bed surface layer was calculated to be 48.8% of the core inventory. Only the surface and near surface Samples 1, 4, and 5 contained appreciable amounts of tellurium. The tellurium content in other samples (Sample 3, and 6 to 11) was low and only up to eleven subsamples contained tellurium per 100 subsamples analyzed.

Although estimates of tellurium retained in the debris bed indicate a large fraction of core inventory (49%) was concentrated in the top surface

layer, the reader is cautioned that there are large uncertainties in these measurements and estimates. Tellurium separation methods as discussed in Appendix A are required to better quantify the tellurium retained in the debris bed.

In addition to tellurium; uranium, zirconium, tin, iron, nickel, chromium, and other elements were also detected in the samples. The atom ratios of elements Sn, U, Zr, Fe, Cr, Ni and tellurium were not consistent.

Calculation of Tellurium Release from the TMI-2 Core

The details of the TMI-2 accident sequence have been discussed in several reports.[22-26] Some of the key events[12] in the accident sequence for the time period 100 to 213 min are shown in Table 4. The critical period of the accident sequence from the point of view of core damage and fission product release is believed to be between 113 and 208 min after the reactor scram.[12] The 113-min time corresponds to the beginning of core uncovery following phase separation in the reactor coolant; the reactor coolant pumps were turned off at about 100 min. The 208 min time corresponds to the core refill following resumption of sustained high pressure injection at about 200 min.

The tellurium release fraction (from the core) was estimated using temperatures calculated with the SCDAP computer code[27] and release rates calculated by the Lorenz model.[2] For these calculations, the core was divided into seven axial and three radial nodes. As shown in Figure 11, the radial nodes are denoted by Cold (C), Average (A) and Hot (H) regions, and the axial nodes were numbered from 1 through 7. The core temperature history and the fractional release rates (estimated from Lorenz's model)[2,7] for the cold, average and hot regions are presented in Table 5.

The SCDAP computer code was used to calculate the extent of zircaloy cladding oxidation.[27] As shown in Figure 11, the cladding in Nodes H6 and A6 were oxidized to $\geq$90%. The rest of the cladding in the core was oxidized to <90%. The estimated fractional releases from the hot, average, and cold regions of the core were 2.2, 6.2, and 1.6%,

TABLE 4. SUMMARY OF PERTINENT EVENTS IN THE TMI-2 ACCIDENT SEQUENCE

Time (min)	Event Number	Event
100	1	Last Reactor Coolant (RC) pumps turned off in Loop A
113	2	Beginning of core uncovery
139	3	Pilot Operated Release Valve (PORV) closed
145	4	Iodine in the reactor building air sample (HP-P-227) began to increase rapidly
150	5	A radiation detector (in core instrument panel area monitor) showed response indicating release of activity to the primary system
174	6	RC Pump 2B was started and ran until 193 min
192	7	The PORV block valve was opened and cycled several times in the next period
200	8	Sustained High Pressure Injection (HPI) and core reflooded
208	9	Core refilled

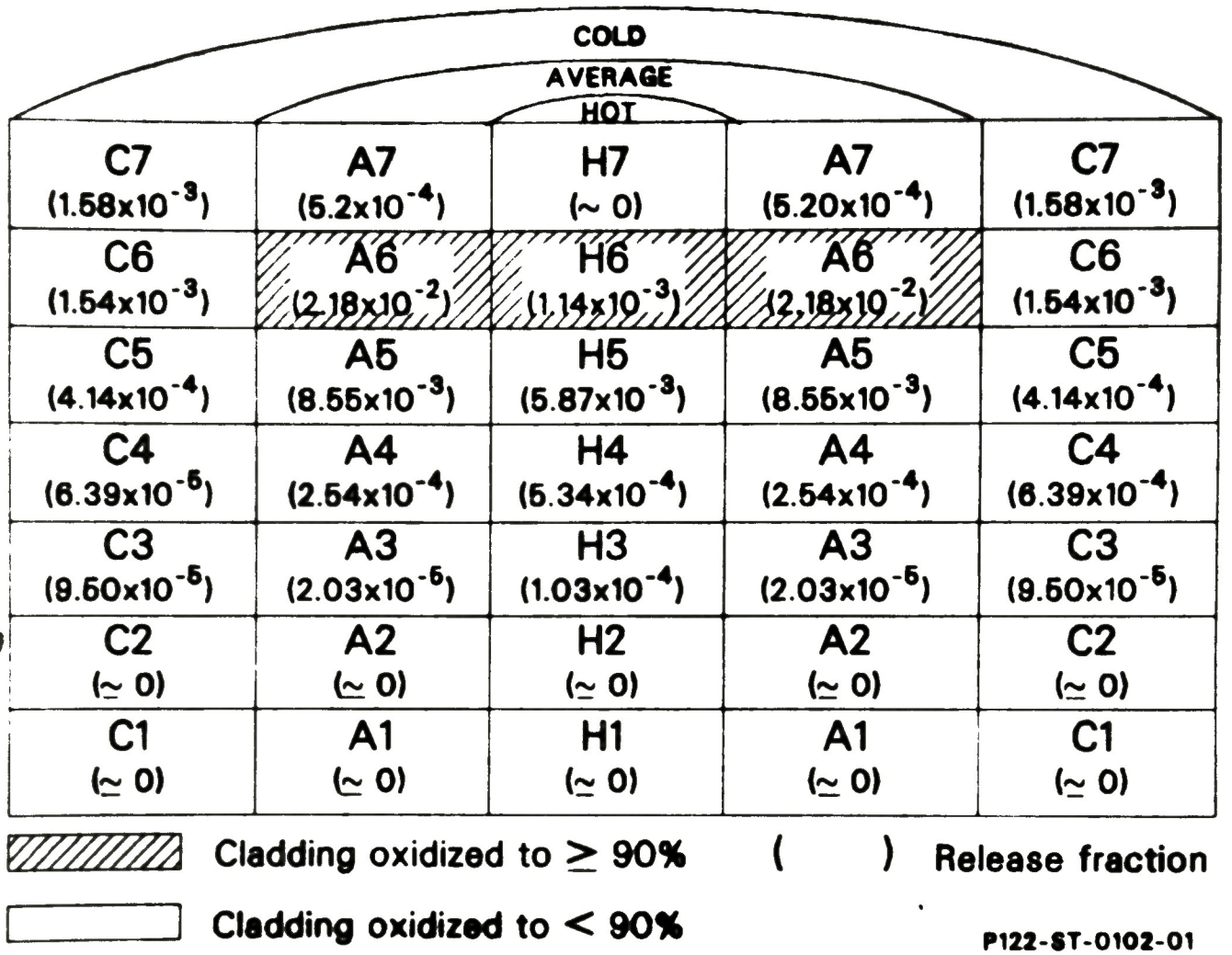

Figure 11. Extent of TMI-2 cladding oxidation and release fractions.

TABLE 5. TMI-2 CORE TEMPERATURE AND FRACTIONAL RELEASE RATE HISTORIES

Time After Trip (min)	Core Temperature[a] (K)					Fractional Release Rate[b] (min^{-1})				
	Node 3	Node 4	Node 5	Node 6	Node 7	Node 3	Node 4	Node 5	Node 6	Node 7
HOT REGION										
140	534	776	923	1036	1024	--	--	--	2.1×10^{-6}	4.7×10^{-8}
145	589	925	1109	1162	1141	--	--	1.2×10^{-7}	8.1×10^{-6}	1.6×10^{-7}
150	746	1096	1305	1271	1254	--	1.0×10^{-7}	9.3×10^{-7}	2.6×10^{-5}	5.4×10^{-7}
155	969	1313	1652	1443	1394	--	1.0×10^{-6}	3.7×10^{-5}	1.6×10^{-4}	2.4×10^{-6}
160	1200	1712	2617	1551	1497	3.0×10^{-7}	6.9×10^{-5}	9.0×10^{-3}	5.0×10^{-4}	5.6×10^{-6}
165	1629	2631	2713	1681	1559	2.9×10^{-5}	9.4×10^{-3}	1.2×10^{-2}	2.0×10^{-3}	1.4×10^{-5}
170	1798	2674	2792	2111	1620	1.7×10^{-4}	1.2×10^{-2}	1.6×10^{-2}	5.3×10^{-2}	2.6×10^{-5}
175	1962	2746	2874	2510	1695	6.1×10^{-4}	2.0×10^{-2}	2.0×10^{-2}	2.6×10^{-1}	5.8×10^{-5}
180	613	612	612	619	756	--	--	--	--	--
185	630	702	749	742	811	--	--	--	--	--
190	811	885	934	833	914	--	--	--	--	--
195	1002	1075	1119	926	1019	--	--	--	--	--
200	1174	1257	1295	1019	1124	--	--	--	--	--
Cladding oxidation	<90%	<90%	<90%	≥90%	<90%	<90%	<90%	<90%	≥90%	<90%
AVERAGE REGION										
140	534	692	861	1013	1001	--	--	--	1.7×10^{-6}	3.7×10^{-8}
145	537	824	1019	1143	1105	--	--	4.4×10^{-8}	6.6×10^{-6}	1.1×10^{-7}
150	674	961	1185	1248	1219	--	--	2.6×10^{-7}	2.0×10^{-5}	3.7×10^{-7}

TABLE 5. (continued)

Time After Trip (min)	Core Temperature[a] (K)					Fractional Release Rate[b] (min^{-1})				
	Node 3	Node 4	Node 5	Node 6	Node 7	Node 3	Node 4	Node 5	Node 6	Node 7
					AVERAGE REGION (continued)					
155	861	1132	1416	1401	1355	--	1.5×10^{-7}	3.0×10^{-6}	1.0×10^{-4}	1.6×10^{-6}
160	1051	1338	1949	1543	1460	6.2×10^{-8}	1.3×10^{-6}	5.7×10^{-4}	4.6×10^{-4}	4.8×10^{-6}
165	1355	1668	2643	1651	1519	1.6×10^{-6}	4.4×10^{-5}	9.7×10^{-3}	1.5×10^{-4}	9.0×10^{-6}
170	1536	1807	2697	1843	1587	1.1×10^{-5}	1.9×10^{-4}	1.2×10^{-2}	1.5×10^{-3}	1.8×10^{-5}
175	1713	1942	2771	2300	1642	7.0×10^{-5}	8.0×10^{-4}	1.5×10^{-2}	1.3×10^{-2}	3.3×10^{-5}
180	612	612	2008	2404	2512	--	--	7.8×10^{-4}	1.4×10^{-2}	6.5×10^{-3}
185	675	761	1960	2413	2517	--	--	6.0×10^{-4}	1.9×10^{-1}	6.6×10^{-3}
190	850	919	2044	2465	2542	--	--	9.4×10^{-4}	2.3×10^{-1}	7.2×10^{-3}
195	1045	1077	2160	2520	2583	--	--	1.7×10^{-3}	2.7×10^{-1}	8.1×10^{-3}
200	1221	1211	2272	2565	2627	--	--	3.1×10^{-3}	3.7×10^{-1}	9.3×10^{-3}
Cladding oxidation	<90%	<90%	<90%	≥90%	<90%	<90%	<90%	<90%	≥90%	<90%
					COLD REGION					
140	534	623	788	929	940	--	--	--	--	--
145	537	750	916	1044	1016	--	--	--	5.8×10^{-8}	4.3×10^{-8}
150	592	875	1053	1145	1071	--	--	6.4×10^{-8}	1.7×10^{-7}	7.7×10^{-7}
155	738	1011	1200	1222	1131	--	4.1×10^{-8}	3.0×10^{-7}	3.8×10^{-7}	1.5×10^{-7}
160	897	1166	1402	1297	1209		2.1×10^{-7}	2.6×10^{-6}	8.5×10^{-7}	3.3×10^{-7}

TABLE 5. (continued)

Time After Trip (min)	Core Temperature[a] (K)					Fractional Release Rate[b] (min^{-1})				
	Node 3	Node 4	Node 5	Node 6	Node 7	Node 3	Node 4	Node 5	Node 6	Node 7
					COLD REGION	(continued)				
165	1052	1353	1748	1364	1266	6.3×10^{-8}	1.5×10^{-6}	1.0×10^{-4}	1.7×10^{-6}	6.1×10^{-7}
170	1215	1643	2426	1410	1315	3.6×10^{-7}	3.3×10^{-5}	5.0×10^{-3}	2.8×10^{-6}	1.0×10^{-6}
175	1413	2247	2635	1456	1391	3.6×10^{-6}	2.7×10^{-3}	9.6×10^{-3}	4.6×10^{-6}	2.3×10^{-6}
180	619	619	2130	2243	1720	--	--	1.5×10^{-3}	2.6×10^{-3}	7.6×10^{-5}
185	613	673	2024	2227	2227	--	--	8.4×10^{-4}	2.4×10^{-3}	2.4×10^{-3}
190	725	789	2071	2270	2255	--	--	1.1×10^{-3}	2.6×10^{-3}	2.8×10^{-3}
195	861	912	2155	2314	2291	--	--	1.7×10^{-3}	3.5×10^{-3}	3.3×10^{-3}
200	994	1032	2240	2347	2315	--	--	2.6×10^{-3}	3.9×10^{-3}	3.5×10^{-3}
Cladding oxidation	<90%	<90%	<90%	<90%	<90%	<90%	<90%	<90%	<90%	<90%

a. Node 3 = 0.943 to 1.386 m, Node 4 = 1.386 to 1.829 m, Node 5 = 1.829 to 2.351 m, Node 6 = 2.351 to 2.874 m, and Node 7 = 2.874 to 3.396 m from the bottom of the core.

b. Fractional release rate from each node. For example 100% of tellurium content in Nodes A6 and H6 is released.

respectively. These estimates were made based on Lorenz's model, and by weighting the core inventory according to the axial flux distribution. The total release from the core is therefore ~10%.

This low estimated tellurium release fraction is consistent with the low measured tellurium release fraction and suggests that most of the tellurium was retained within the core.

TRAP-MELT Calculations

TRAP-MELT[28] is a dynamic numerical model which calculates fission product particle and vapor transport and deposition in LWR primary systems during meltdown accidents. The transport and retention of fission products within the primary coolant system during a meltdown accident are treated in terms of control volumes and flow connections. It assumes that fission product transport can be superimposed on the fluid flow without coupling to it. A radionuclide species can reside within a control volume in two states, particle and vapor form. The geometry of the system, mass generation rates of a limited number of fission products (iodine, cesium, and tellurium) time-dependent thermal-hydraulic data, and radionuclide physical properties[a] are required as input parameters for the code. The code calculates the transport and deposition of these radionuclides in the control volumes as a function of time.

Preliminary calculations of tellurium transport and deposition during the TMI-2 accident were made using the TRAP-MELT computer code. Input parameters were obtained from various TMI-2 reports published[12,24,26,29] during the last five years. The primary coolant system was divided into eight control volumes as shown in Figure 12. The control volume geometries were obtained either from the Final Safety Analysis Report (FSAR)[30] or estimated. These parameters include length, hydraulic diameter, flow area, settling area, and height. Sixteen 5 min time intervals starting from 140 min and ending with 220 min were used. Steam temperatures and steam flow rates reported in Reference 26 were used and the system pressures were

a. Average radii and particle density of fission products.

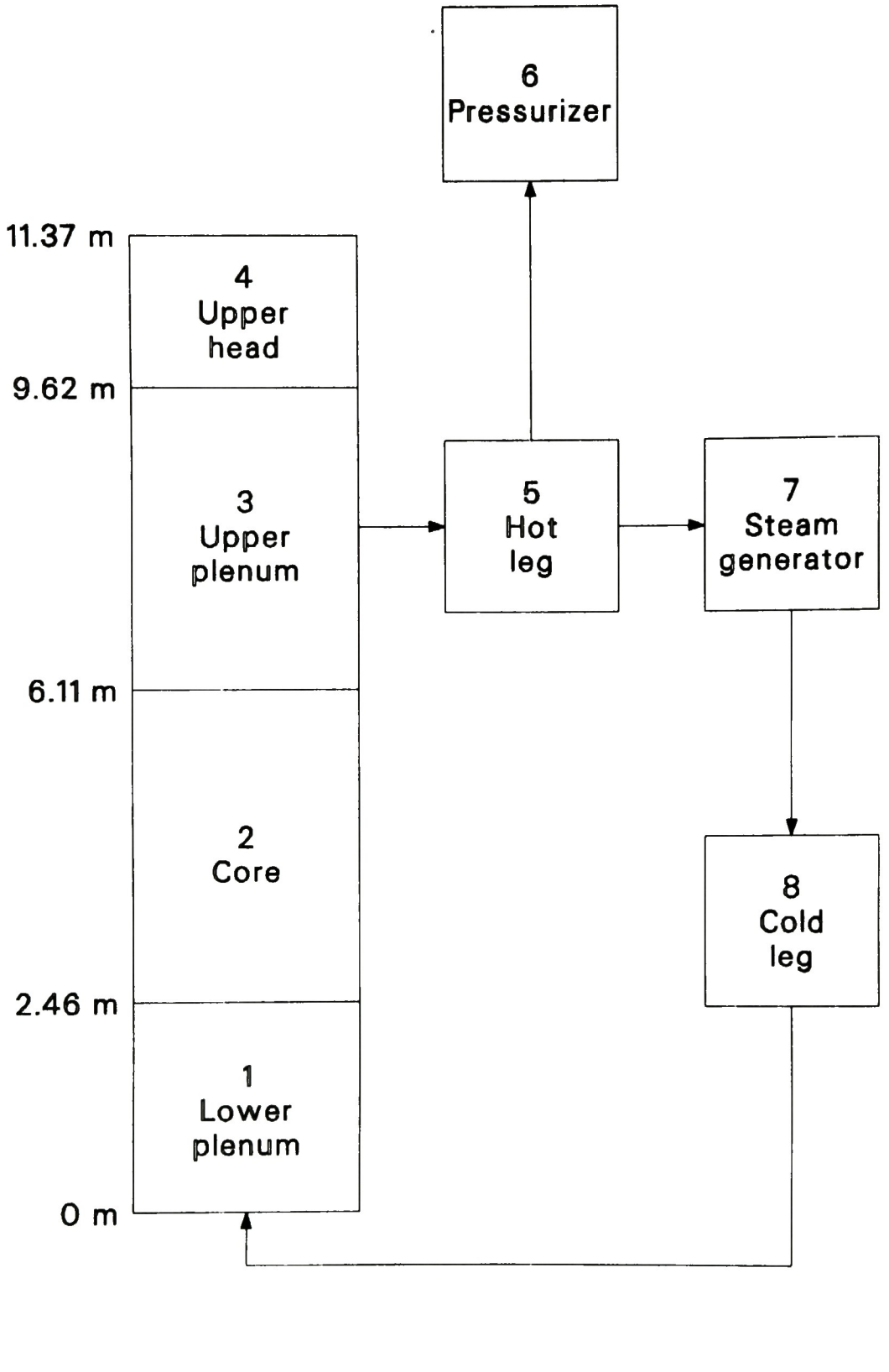

Figure 12. TMI-2 control volumes for TRAP-MELT calculations.

obtained from the measured charts reported in Reference 29. The tellurium source term (10%) estimated in the previous section was used. TRAP-MELT input parameters are presented in Appendix B.

The TRAP-MELT predicted fraction of the core inventory of tellurium deposited on various control volume (Figure 12) surfaces versus time are shown in Figures 13 through 19. The fraction of the core inventory deposited on (a) lower plenum, (b) core, (c) upper plenum, (d) upper head, (e) hot leg, (f) pressurizer, (g) steam generator, and (h) cold leg surfaces was predicted to be 1.90×10^{-4}, 0.0, 5.4, 1.7, 0.8, 0.1, 0.1, and 0.003%, respectively. Of a total deposition of 5.4% on upper plenum surfaces, about 2.9% was predicted to be deposited after the event at 200 min, when the core was reflooded (see Table 4 for accident sequence). The large steam flow rate [26] at reflood was the principal reason for this calculated result. The measured tellurium deposition on the upper plenum surfaces was 1.8%. The TRAP-MELT calculated tellurium deposition was 5.4%.

Review of ORNL Tellurium Release Results

An extensive review of available tellurium release data was performed by Lorenz et al.[2] at ORNL. This examination of the existing tellurium release data indicates both high and low releases. High releases were observed in tests that contained no zircaloy cladding or highly oxidized ($\geq$90% conversion to ZrO_2) zircaloy. Low releases were observed in tests that contained zircaloy cladding with a lower extent of oxidation (<90%). Tellurium release data from three high temperature fission product release tests (HI series tests) are presented in Table 6. In the high temperature Tests HI-1, HI-2, and HI-3, high burnup commercial pressurized water reactor (PWR) fuel from the H. B. Robinson reactor was heated in steam at maximum temperatures of 1673, 1973, and 2273 K, respectively. Tellurium released from the 20.32-cm long fuel rod segments was measured by spark source mass spectrometry. In Test HI-2, oxidation of the zircaloy cladding was complete and a larger amount of tellurium was released than in the other tests in which oxidation was limited. Lorenz et al.[2] have proposed a tellurium release rate model which is based on

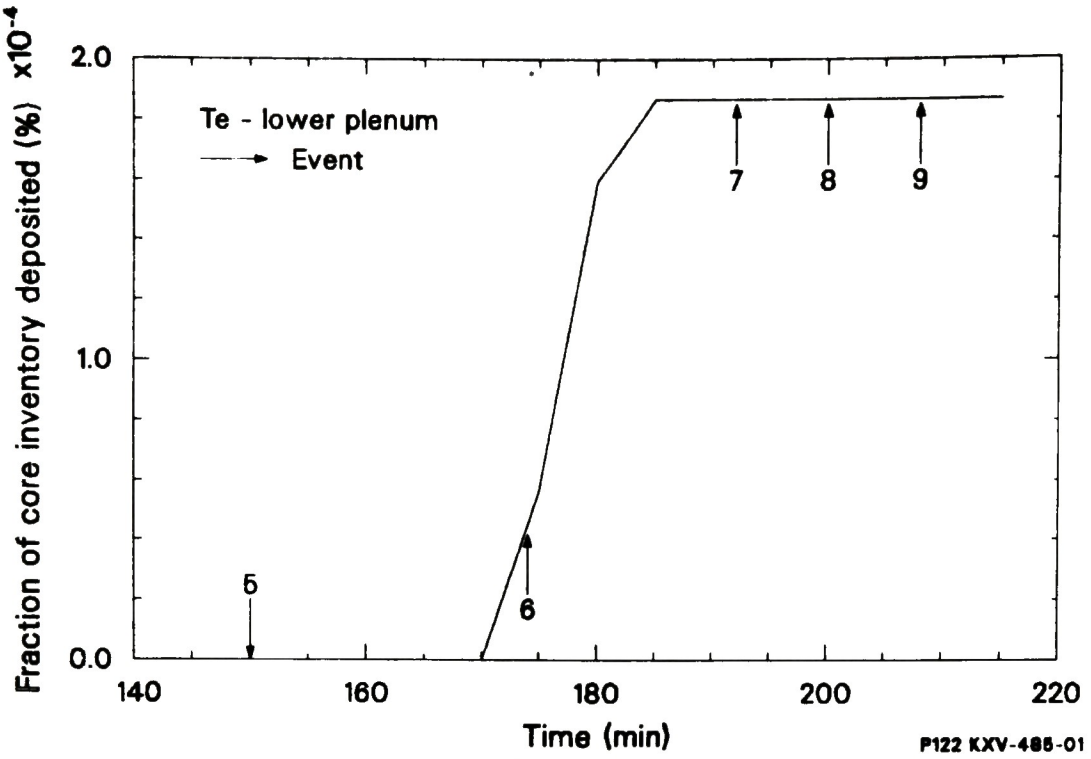

Figure 13. TRAP-MELT predicted tellurium deposition on TMI-2 lower plenum surfaces.

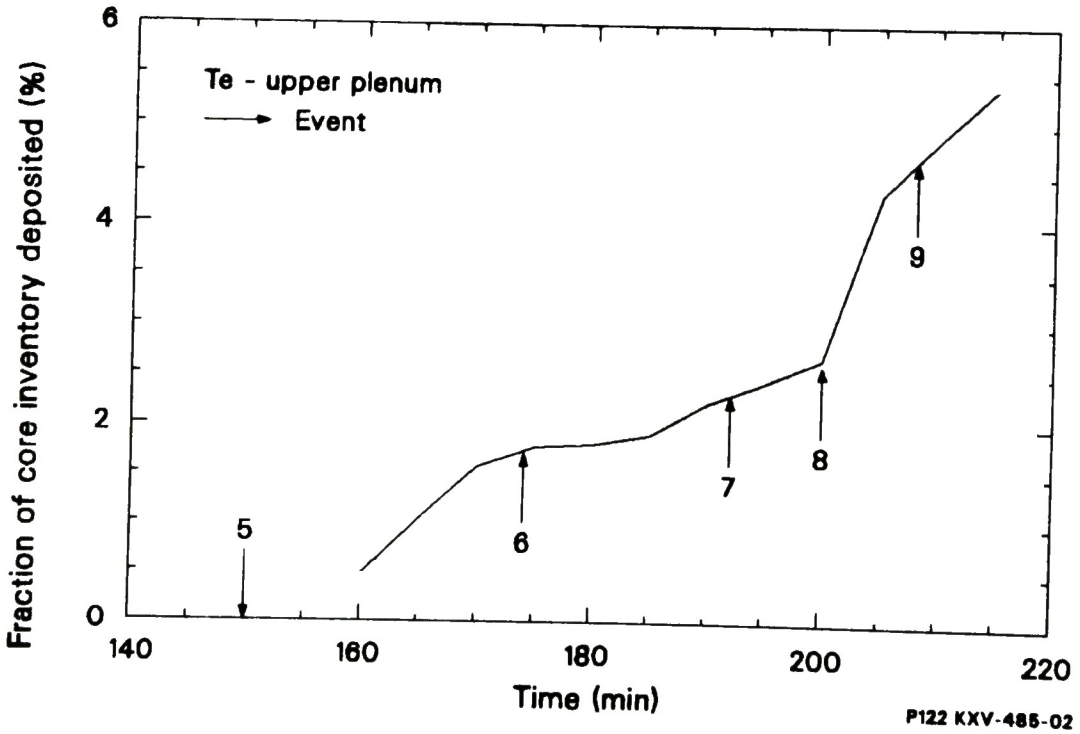

Figure 14. TRAP-MELT predicted tellurium deposition on TMI-2 upper plenum surfaces.

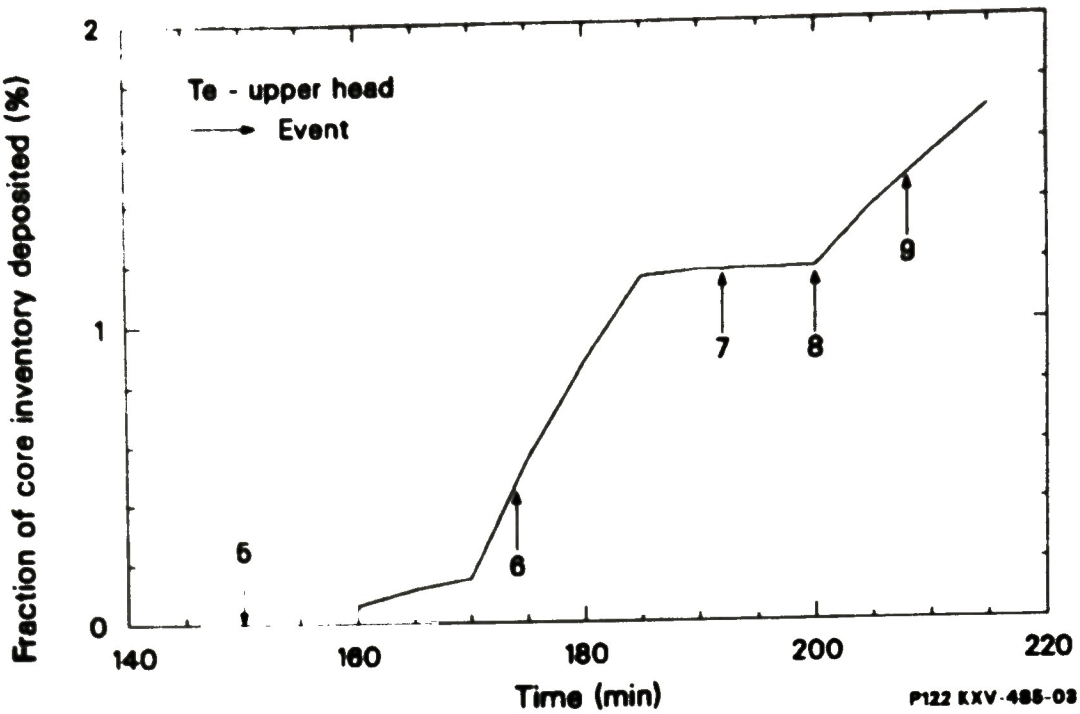

Figure 15. TRAP-MELT predicted tellurium deposition on TMI-2 upper head surfaces.

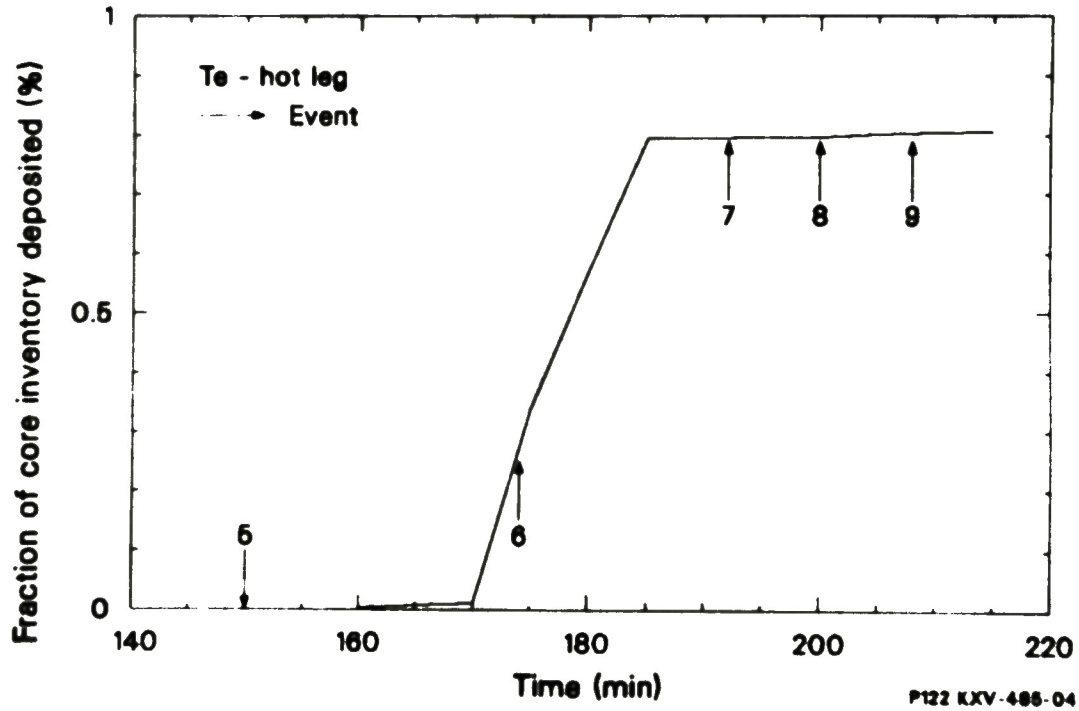

Figure 16. TRAP-MELT predicted tellurium deposition on TMI-2 hot leg surface.

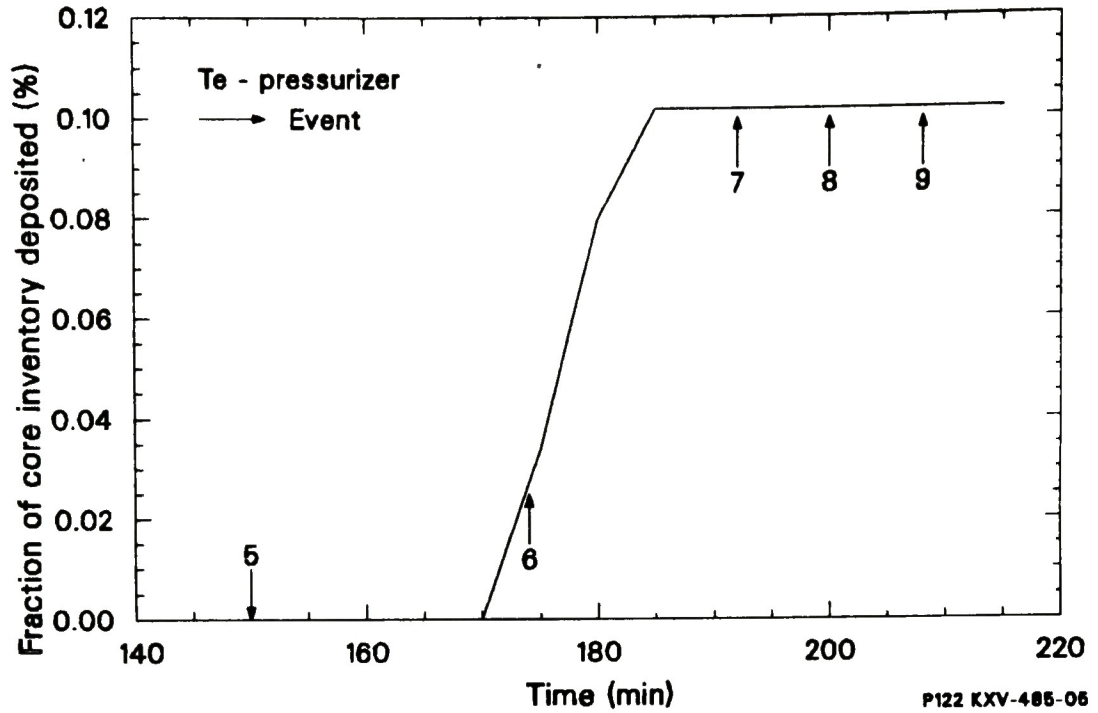

Figure 17. TRAP-MELT predicted tellurium deposition on TMI-2 pressurizer surface.

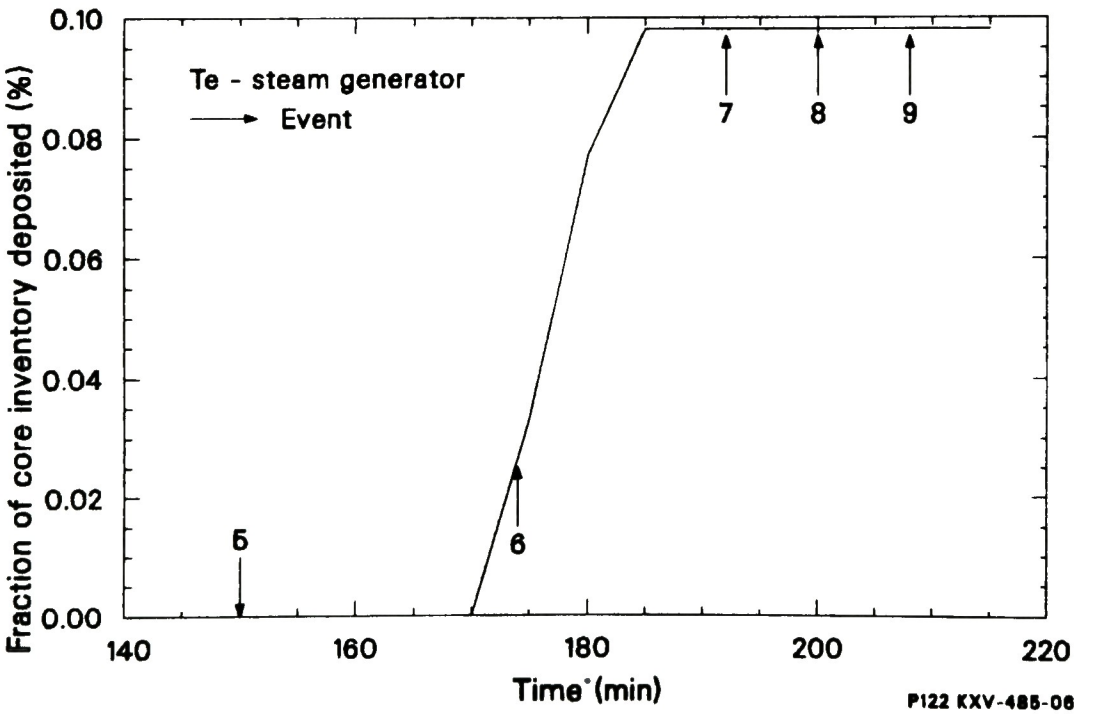

Figure 18. TRAP-MELT predicted tellurium deposition on TMI-2 steam generator surfaces.

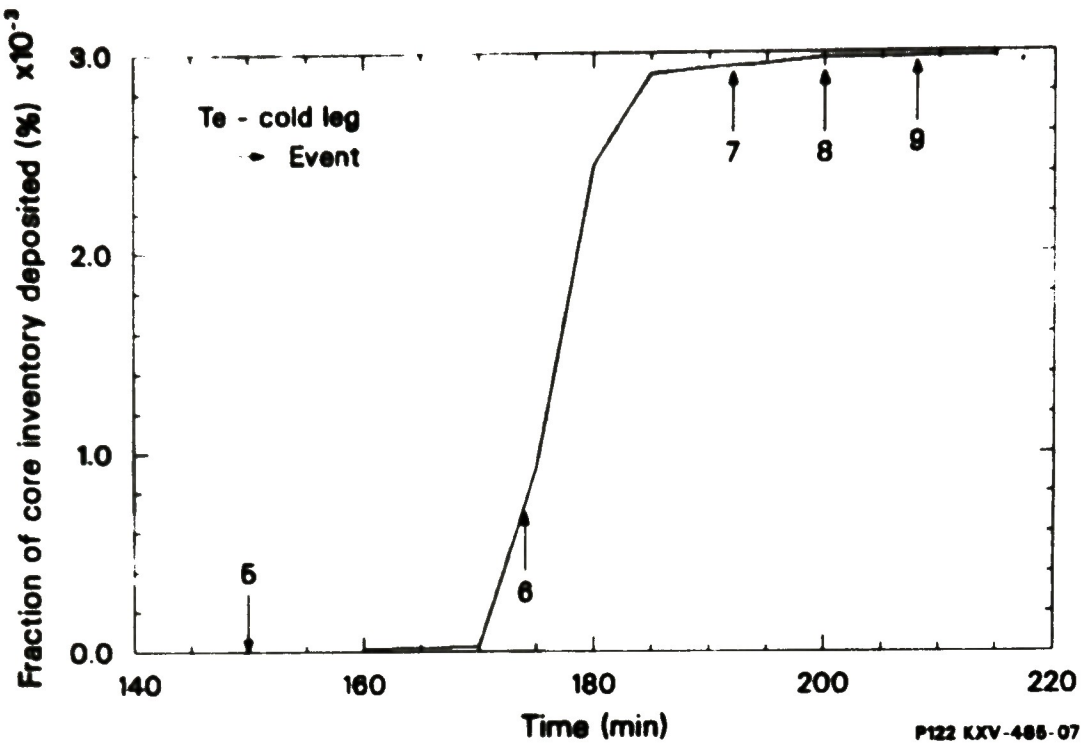

Figure 19. TRAP-MELT predicted tellurium deposition on TMI-2 cold leg surface.

TABLE 6. TELLURIUM RELEASE IN HI SERIES TEST AT ORNL

Test	Temperature (K)	Time (min)	Zircaloy Oxidized (%)	Te Release (%)	Te Fractional Release Rate (fraction/minute)
HI-1	1673	30	40	0.3	7.0×10^{-5}
HI-2	1973	20	100	50 to 100	6.0×10^{-2}
HI-3	2273	20	35	0.6	2.4×10^{-4}

the extent of zircaloy oxidation. The fractional release rate, k(T) in fraction per minute, is obtained from the following expression:

$$k(T) = A\, e^{B(T-273)} \qquad (2)$$

where A and B are constants given in Table 7, and T is temperature in K. When the local degree of cladding oxidation is <90%, a low release rate is suggested. When the local degree of cladding oxidation is $\geq$90%, a high release rate is suggested. Lorenz et al. indicate that the threshold for change in release rate is a function of local oxidation and that the core average oxidation is not an acceptable basis. This model was used to estimate the tellurium release of 10% during the TMI-2 accident in the previous section.

Kelly et al.,[31], have suggested that the fractional release rate should exhibit the usual Arrhenius temperature dependence of the form

$$K = K_0 \exp(-Q/RT) \qquad (3)$$

instead of the A exp (BT) used in NUREG-0772,

where

Q = activation energy in kcal/mol

R = gas constant in 1.986 cal/mol.K and

T = temperature in K.

The constants Q and K_0 are given in Table 8 for both cases of cladding oxidation <90% and $\geq$90%. The estimated factional releases from the hot, average and cold regions of the core were 3.2, 8.6, and 2.2%, respectively. The total release from the core is therefore 14.0%.

The highlights of the results of three recent ORNL control tests,[32,33] C-7, C-8 and C-9 are presented here. The Tests C-7 and C-8

TABLE 7. COEFFICIENTS FOR LORENZ TELLURIUM RELEASE RATE MODEL

Cladding Condition	Temperature (K)	A	B
Zircaloy oxidation <90%	<1873	1.625×10^{11}	1.061×10^{-2}
	1873 to 2273	9.04×10^{-8}	5.22×10^{-3}
	>2273	6.025×10^{-6}	3.12×10^{-3}
Zircaloy oxidation ≥90%	<1873	6.50×10^{-10}	1.061×10^{-2}
	1873 to 2273	3.616×10^{-6}	5.22×10^{-3}
	>2273	2.41×10^{-4}	3.12×10^{-3}

TABLE 8. PARAMETERS FOR ARRHENIUS TEMPERATURE DEPENDENCE RELEASE RATE MODEL

Cladding Condition	$Q\left(\frac{kcal}{mol}\right)$	K_o (min^{-1})
Zircaloy oxidation <90%	55.74	9.015×10^{2}
Zircaloy oxidation ≥90%	53.18	3.155×10^{4}

were conducted to examine the deposition behavior of CsOH, CsI and Te in an experimental apparatus containing platinum and stainless steel thermal gradient tubes (TGT), respectively. A long section (5.0 cm) of the TGT at the inlet end in the C-8 test was made of Inconel 600. In control Tests C-7 and C-8, traced CsOH, CsI and Te species were vaporized and transported by a steam-hydrogen carrier gas through a zirconia liner to the collection system. In these tests hot zones in the zirconia liner were maintained at 1073 and 1273 K, respectively.

In control Test C-7, 88% of tellurium was released and transported to the collection system. About 86% was deposited in the platinum TGT. Of that, 14% dissolved in the platinum. A large fraction (80%) of the tellurium reacted with CsOH in the gas phase and was found in the surface deposit of the Platinum TGT portion at 973 K. The identified reaction product was CsTe.

In Test C-8, about 52% of tellurium was released and carried to the collection train. About 51.7% was deposited in the stainless steel TGT. Of the tellurium that entered the TGT about 70% was deposited in the Inconel-600 section at the inlet end of the TGT, which was at 1073 to 1200 K. The likely reaction products are nickel and chromium tellurides.

In Test C-9 an irradiated tellurium specimen[a] was placed in a 15.24 cm long PWR type fuel specimen. The traced tellurium was placed in a ZrO_2 tube at the inlet end of the fuel specimen. The rest of Zircaloy-4 cladding space was filled with UO_2 pellets. Zircaloy end caps were used to close the ends of the cladding. A 1.58 mm hole was drilled through the outlet end of the cladding. A platinum TGT was used and the traced ^{129m}Te was vaporized and transported by helium-steam carrier gas through the platinum TGT to the collection system. The temperature of the test specimen was maintained at 1973 K during the test.

In Test C-9, about 97% of the tellurium was released. The largest fraction (~57%) was found in the platinum TGT. Almost all of the

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a. 39.8 mg of metallic tellurium irradiated to obtain 31 mCi of ^{129m}Te activity.

tellurium was released when the zircaloy cladding oxidation was near completion. Most of the tellurium that was collected in the TGT deposited at 873 K. Examination of the surface deposit revealed that tin and tellurium were present in a 1:1 atom ratio. It is proposed that the tellurium is retained by zircaloy cladding either by reacting with zirconium to form zirconium telluride, or by dissolving in the zircaloy. Once the zirconium is converted to ZrO_2 by steam oxidation, the zircaloy cladding released tellurium. The liberated tellurium contacted the liquid tin droplets that are dispersed in the oxidized cladding, reacted and formed SnTe and was released in that chemical form. Tellurium release data from these three control tests are presented in Table 9. The estimated tellurium release rates from C-7, C-8, and C-9 were 1.1×10^{-3}, 2.3×10^{-3} and 2.6×10^{-2} (min^{-1}), respectively. The zircaloy cladding in Test C-9 was completely oxidized.

Tellurium Release from PBF Severe Fuel Damage Tests

A series of Severe Fuel Damage (SFD) tests is being conducted by EG&G Idaho Inc., in the Power Burst Facility (PBF) at the Idaho National Engineering Laboratory for the U.S. Nuclear Regulatory Commission and their international sponsors.[a] [34,35]

A major objective of these tests is to measure the release, transport, and deposition of fission products during in-pile tests similar to the accident which occurred at the TMI-2 reactor. Four bundle experiments have been completed in the SFD program. Each bundle is operated at full power for three days, and then cooled for at least eight days to build up an appropriate ratio of cesium to iodine. Shortly before the transient, the bundle is irradiated for four hours to build up an adequate inventory of short-lived fission products. The inlet flow to the bundle is then reduced and the bundle power is increased to initiate the transient portion of the experiments, and to force the water level to decrease, leaving the bundle to be cooled by steam. The bundle heats up in superheated steam to peak

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a. Sponsors of the program include Belgium, Canada, Federal Republic of Germany, Italy, Japan, Netherlands, Republic of China (Taiwan), Republic of Korea, Sweden, United Kingdom, and United States.

TABLE 9. TELLURIUM RELEASE IN CONTROL TESTS AT ORNL

Test	Maximum Temperature (K)	Time At Maximum Temperature (min)	TGT Material	Te Release %	Te Fractional Release Rate (fraction/minute)	Species Identified on TGT
C-7[a]	1073	31.3	Platinum	88	1.1×10^{-3}	CsTe
C-8[a]	1273	26.3	Stainless Steel	52	2.3×10^{-3}	$NiTe_{0.07}$
C-9[b]	1973	87.5	Platinum	97	2.6×10^{-2}[c]	SnTe

a. CsI, CsOH and Te species were vaporized and transported by a steam-hellium-hydrogen carrier gas through a zirconia liner.

b. Traced tellurium species was vaporized and transported by helium-steam carrier gas through zircaloy clad fuel element.

c. The fractional release rate was estimated from the data when the zircaloy cladding was completely oxidized.

temperatures of about 2400 K. Parameters varied during the four tests are: heatup rate (amount of cladding oxidation); inlet flow (hydrogen to oxygen ratio); cooldown rate (amount of fragmentation); test rod burnup; and presence of control material. Unirradiated test fuel rods were used during the first two tests, and irradiated fuel rods (36 GWd/tU) were used in the third and fourth tests, with control rods included in the fourth test. The SFD test program is summarized in Table 10. The SFD scoping test (SFD-ST), Tests SFD 1-1, 1-3 and 1-4 were completed on October 29, 1982, September 8, 1983, August 3, 1984, and February 7, 1985, respectively. Data from the SFD-ST, SFD 1-1 and SFD 1-3 are presented below.

The test trains of the SFD scoping test and Test SFD 1-1 contained a 32 rod bundle of 0.91-m-long 17 x 17 pressurized-water-reactor type fresh fuel rods (6.2 wt% enriched with ^{235}U). The test trains of SFD 1-3 and SFU 1-4 contained a bundle of irradiated rods and two fresh rods. In addition, the test trains of SFD 1-3 and SFD 1-4 each contained four zircaloy guide tubes and Test SFD 1-4 contained four Ag-In-Cd control rods. The entire bundle of each test was enclosed in an insulated shroud as shown in Figure 20. During the transient, coolant entered the bottom of the fuel bundle, passed through the bundle, and then exited through an effluent steamline which connected to the sampling and monitoring system. Fuel behavior was monitored during the tests with cladding surface, fuel centerline and shroud thermocouples, flow meters, steam probes, and other instrumentation.

During the SFD-scoping test, the bundle was subjected to a slow heatup (0.13 K/s) to 1700 K, and then to rapid heating (~10 K/s) to 2400 K, followed by a rapid quench. During the second, third, and fourth tests (SFD 1-1, SFD 1-3, and SFD 1-4), the test bundle was subjected to a more rapid heating (~0.45 K/s) to 1300 K, 1.3 K/s to 1700 K and then more rapid heating (~30 K/s) to 2400 K. The SFD 1-1, SFD 1-3, and SFD 1-4, were slow-cooled rather than quenched. Considerable cladding oxidation (virtually all zircaloy in the test bundle), cladding melting, fuel liquefaction (18% of the bundle), and fuel fragmentation (60% of the bundle) occurred during the SFD-ST. Limited cladding oxidation (30%) and significant cladding melting and fuel liquefaction occurred during Test

TABLE 10. PBF SEVERE FUEL DAMAGE TESTS

Test	Specified Heating Rate (K/s)[a]	Inlet Flow (g/s)	Test Bundle	Cool Down	Completion Date
SFD-ST	0.16	16.4	32 Fresh rods	Quench	10/29/82
SFD 1-1	0.44 to 1300 K 1.3 to 1700 K	0.6	32 Fresh rods	Slow	9/08/83
SFD 1-3	0.44 to 1300 K 1.3 to 1700 K	0.6	26 irradiated rods 2 fresh rods 4 guide tubes	Slow, argon	8/03/84
SFD 1-4	0.44 to 1300 K 1.3 to 1700 K	0.6	26 irradiated rods 2 fresh rods 4 Ag-In-Cd control rods in guide tubes	Slow, argon	2/07/85

a. About 1700 K heating rate is driven by zircaloy oxidation and is typically >10 K/s.

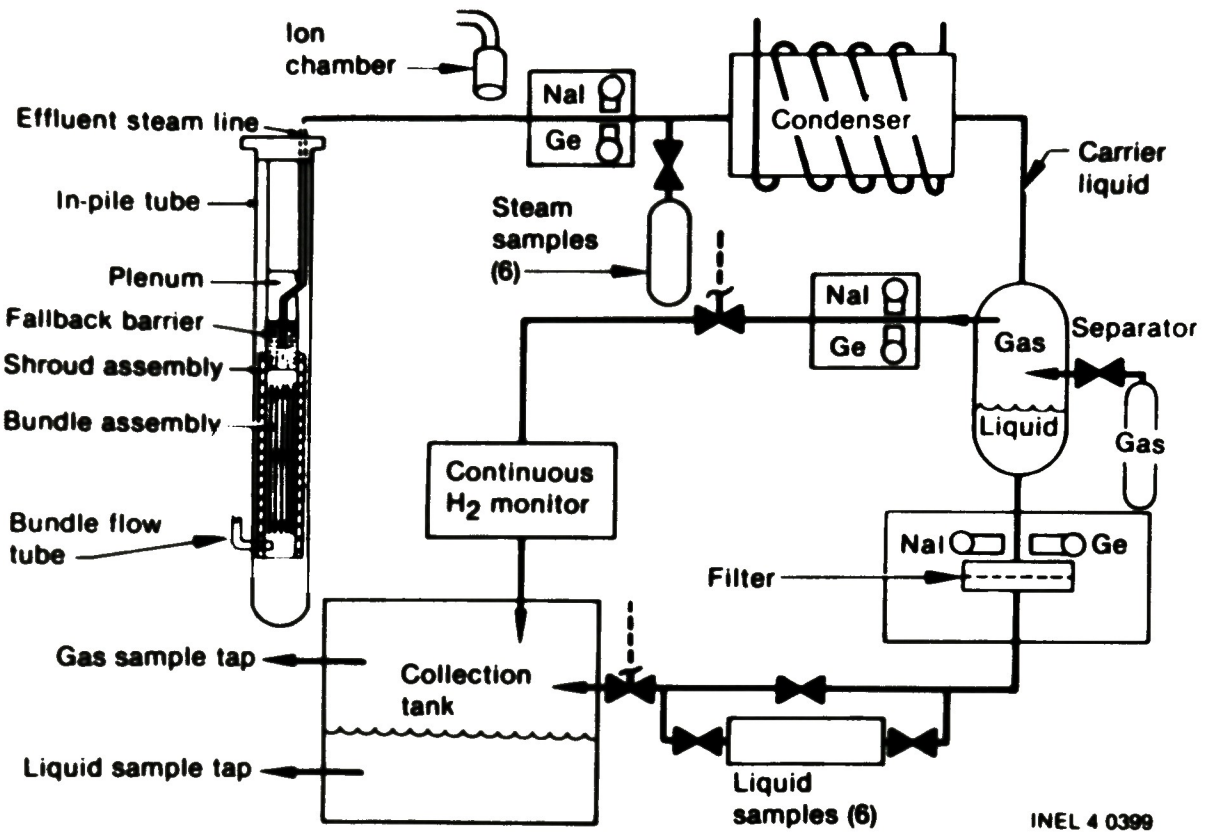

Figure 20. SFD fission product monitoring system.

SFD 1-1. The degree of cladding oxidation in Tests SFD 1-3 and SFD 1-4 had not been analyzed at the time of this report. The SFD 1-3 and SFD 1-4 assemblies used irradiated rods at average burnups from 35 to 37 GWd/t. These tests were generally steam starved experiments. Limited oxidation and extensive liquefaction is expected.

Effluent samples were collected during the SFD tests and analyzed by a variety of analytical techniques.[36,37] The types of samples include: (a) steam samples (filtered and unfiltered), (b) liquid grab samples, (c) filter debris samples, (d) several steamline pipe samples, and (e) liquid and gas samples from the collection tank (see Figure 20).

The fractional release rates (fraction of the bundle inventory released per minute) were calculated from the gas and liquid grab samples.[36] The data for tellurium are presented in Table 11.

The high tellurium release rate measured in SFD-ST, compared to SFD 1-1, is probably due to the extensive oxidation of cladding, the high steam flow rate and the steam plentiful conditions that existed during the SFD-ST. In SFD 1-1, the zircaloy cladding may have retained tellurium because the extent of cladding oxidation was so low; plateout of the tellurium may have occurred in the steamlines due to much slower flow rates.

The distribution (mass balance) of tellurium in various components of the system is presented in Table 12. About 40% of the core inventory of ^{129m}Te was released during SFD-ST; in contrast, <1% was released in SFD 1-1. The principal reason for this differences is believed to be holdup of tellurium by zircaloy in SFD 1-1.

The tellurium release fraction measured in the collection tank liquid for Test SFD 1-3 was 1.4×10^{-4}. The total Te release fraction is expected to be lower than that measured during Test SFD 1-1, because the zircaloy oxidation in SFD 1-3 was less than that of SFD 1-1. Tellurium is

TABLE 11. FRACTIONAL RELEASE RATES OF TELLURIUM FROM PBF SFD TESTS (fraction/minute)

Test	Heatup		Quench (600 K)
	2000 K	2400 K	
SFD-ST	7.0×10^{-6}	5.42×10^{-3}	2.9×10^{-3}
SFD 1-1	3.0×10^{-8}	--	3.8×10^{-5}
SFD 1-3	--[a]	--[a]	--[a]
SFD 1-4	--[a]	--[a]	--[a]

a. Not completed at this time.

TABLE 12. OVERALL MASS BALANCE OF ^{129m}Te IN PBF SFD TESTS (fraction of total bundle inventory)

Samples	Tests			
	SFD-ST[a]	SFD 1-1[b]	SFD 1-3	SFD 1-4
304 SS Steam Lines	1.02×10^{-1}	5.30×10^{-3}	--	--[c]
Grab Samples	7.60×10^{-5}	5.10×10^{-7}	--	--[c]
5 μm 304 SS Filter	1.96×10^{-1}	6.10×10^{-4}	--	--[c]
Collection Tank	9.80×10^{-2}	3.10×10^{-3}	1.4×10^{-4}	--
Total Release Fraction	3.96×10^{-1}	9.01×10^{-3}	--	--[c]

a. Slow heatup and quench, high steam flow rate (16.4 g/s), steam plentiful, and 100% cladding oxidation.

b. Fast heatup, slow cooldown, low steam flow rate (0.6 g/s), steam starved, and 30% cladding oxidation.

c. Analysis not completed at this time.

trapped by the unoxidized zircaloy, consequently one would expect comparable or lower tellurium release fractions in SFD 1-3 than in SFD 1-1. Information about tellurium behavior in Test SFD 1-4 is not yet available.

Tellurium Release from SASCHA Tests

Experiments have been performed[37-39] in the SASCHA facility in Karlsruhe, West Germany, to investigate fission product release under severe damage and core meltdown conditions. The major objectives were (a) to determine the release fractions of radiologically important fission products in the temperature range 1773 to 3073 K (1500 to 2800°C), and (b) to characterize the physical and chemical behavior of the released material. The SASCHA test facility consists of a high frequency induction furnace, a crucible, and devices for aerosol collection and analysis. The fuel rods were composed of zircaloy cladding and UO_2 pellets with a simulated burnup of 44 GWd/t. Stainless steel and absorber materials were included to obtain a representative core melt composition. The intergal melt mass was about 200 to 250 g. The released material was collected on glass fiber filters. The filters and the walls of the transport tube were analyzed.

Tests were conducted in air and in steam. The flow rate and pressure were 10 to 30 L/min and 0.21 MPa (two bars), respectively. The fractional release rates (fraction of the inventory released per minute) are presented in Table 13. The fractional release rates in air and steam were essentially the same. During these early tests, the heating rates were normally $\leq$473 K/min, the zircaloy cladding was almost completely oxidized, and the total estimated tellurium release fraction was 0.81.

Experiments were also conducted with a reduced steam supply (28 L/min Ar 1.5 L/min steam) and with a purely reducing atmosphere (28 L/min Ar 1.5 L/min of H_2). The release fractions are shown in Figures 21 and 22. Lower tellurium release was measured for a reduced steam supply. Although

TABLE 13. FRACTIONAL RELEASE RATES FROM SASCHA TESTS IN AIR AND STEAM (fraction/minute)

Temperature (K)	2073	2173	2273	2473	2673
Air[a]	4.8×10^{-3}	--	--	3×10^{-2}	7.3×10^{-2}
Steam[a]	--	1.8×10^{-2}	--	--	7.1×10^{-2}
Ar + 5% H_2	--	1.8×10^{-3}	--	--	--

a. Flow rate = 10 to 30 L/min, Pressure = 0.2 MPa (2 bars).

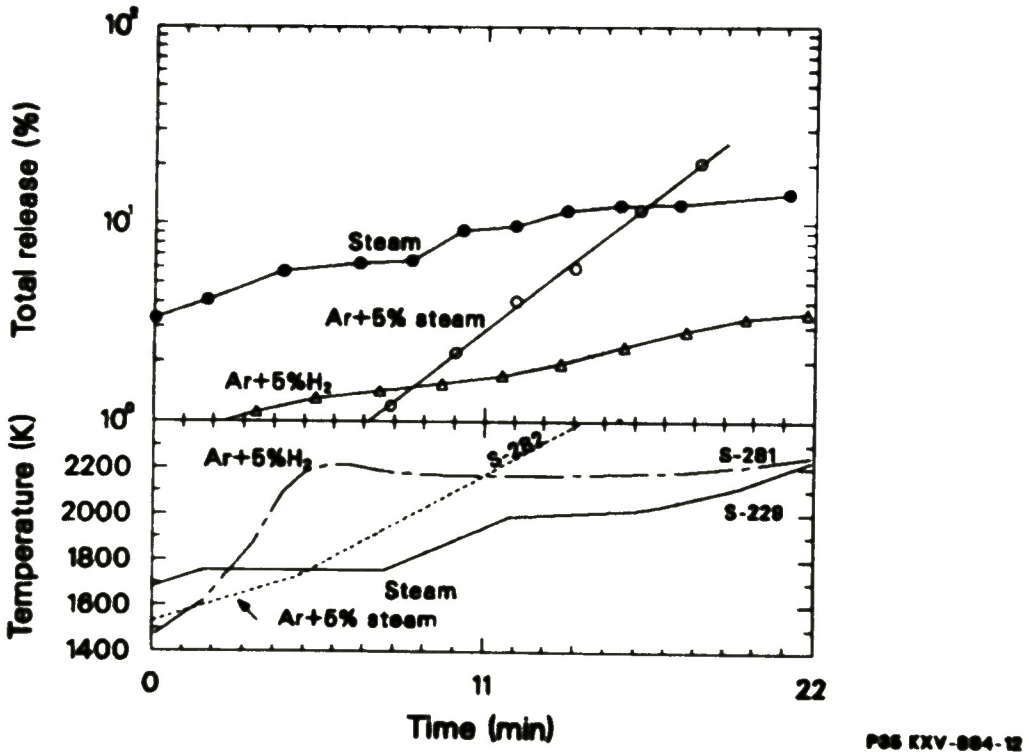

Figure 21. Dependence of tellurium release on steam supply in SASCHA tests.

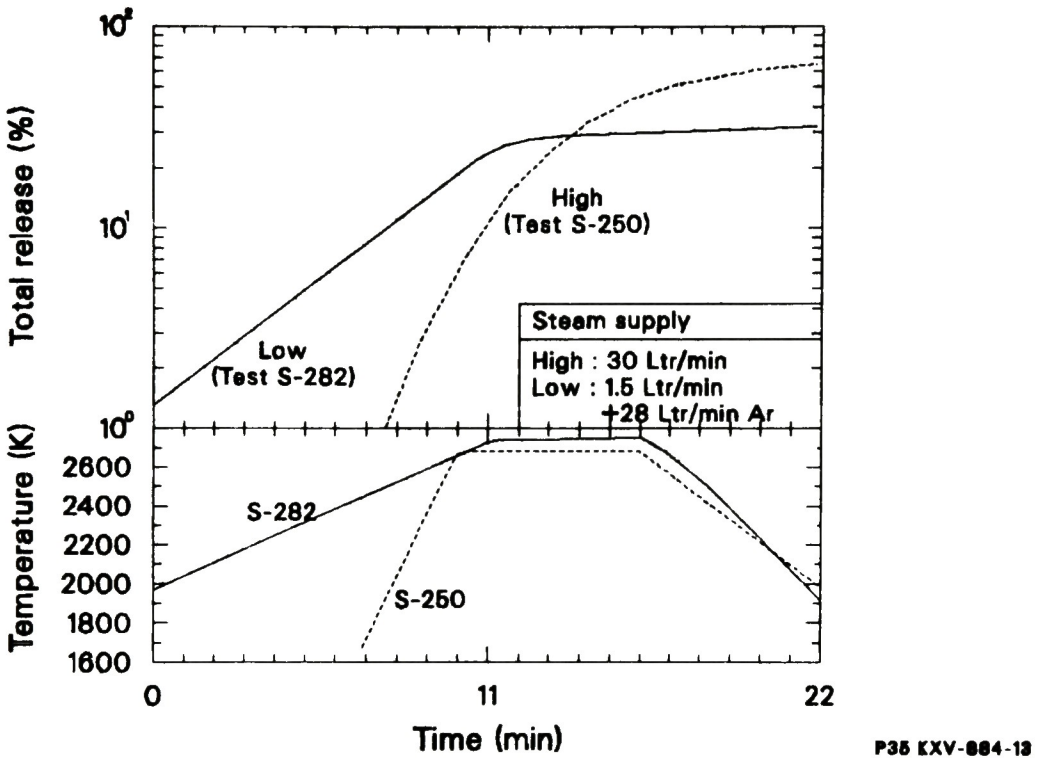

Figure 22. Tellurium release in SASCHA tests with high and low steam supply.

the temperature in Test S-281 (Ar + 5% steam) was 200 to 400 K higher for some minutes, the tellurium release in Test S-229 (steam) about a factor five lower. In Test S-282 (Ar + 5% steam) a low steam supply resulted; the difference in tellurium release between these two cases has been discussed above. Tellurium releases in high and low steam supply tests are compared in Figure 22. Tellurium release in low steam supply test, S-282 (1.5 L/min steam + 28 L/min Ar), was about 30% and in the high steam supply test, S-250 (30 Lmin steam) was about 70%. Albrecht and Wild[38] suggest that the low degree of oxidation causes tellurium to be retained in the melting crucible due to chemical reactions with the zircaloy cladding and (probably) with the stainless steel components. As the oxidation increases, the resulting tellurides are destroyed in favor of metal oxide formation which, in turn, accelerates the release of tellurium.

COMPARISON OF TMI-2 TELLURIUM BEHAVIOR WITH IN-PILE AND OUT-OF-PILE TESTS

In this section, the fractional release rates and release fractions estimated and measured during the TMI-2 accident are compared with measurements from the two PBF Severe Fuel Damage Tests, and the ORNL and SASCHA out-of-pile tests. The modified tellurium release model of Lorenz et al. was used to estimate the fractional release rates for TMI-2 in the temperatures range of 1300 to 2550 K for two regions in the core: 3.05 to 3.66 m (10 to 12 ft) and 2.44 to 3.05 m (8 to 10 ft) from the bottom of the core, where the cladding oxidation was $\geq$90 and <90%, respectively. The fractional release rates versus temperature are shown in Figure 23, where they are compared with the data from the PBF tests (SFD-ST and SFD 1-1), ORNL tests (HI-1, HI-2, HI-3, and C-9) and the SASCHA tests. The results of the PBF SFD-scoping test (at 2400 K), the ORNL Test HI-1 and the SASCHA tests lie above the lower line calculated for TMI-2 with the Lorenz model for <90% cladding oxidation. However, they are below the upper line calculated for $\geq$90% oxidation. The results of the SFD-ST (at 2000 K), the SFD 1-1, and the ORNL Test HI-3 show low tellurium release rates below the $\geq$90% oxidation curve. These results are probably indicative of low zircaloy cladding oxidation in the tests and holdup of tellurium by zircaloy. The release rate measured in Test HI-2 is in reasonable agreement with the curve calculated for cladding oxidation $\geq$90%.

The release fractions measured and estimated from TMI-2 are compared with the in-pile and out-of-pile tests in Table 14. The calculated and measured tellurium release fractions for TMI-2 were low. The measured tellurium fraction in the TMI-2 accident simulation test (SFD 1-1) in the PBF was even lower. The PBF SFD 1-1 test closely approximated the thermal hydraulic conditions of the TMI-2 accident, and the results of this test indicate very small tellurium release ($\sim$0.9%). The SFD 1-1 result is consistent with ORNL tests, where the cladding oxidation was low and tellurium was tied up with the zircaloy cladding. Also in SASCHA tests, the tellurium release was higher in a test where the steam flow was high. The availability of unoxidized zircaloy in the TMI-2 core during the

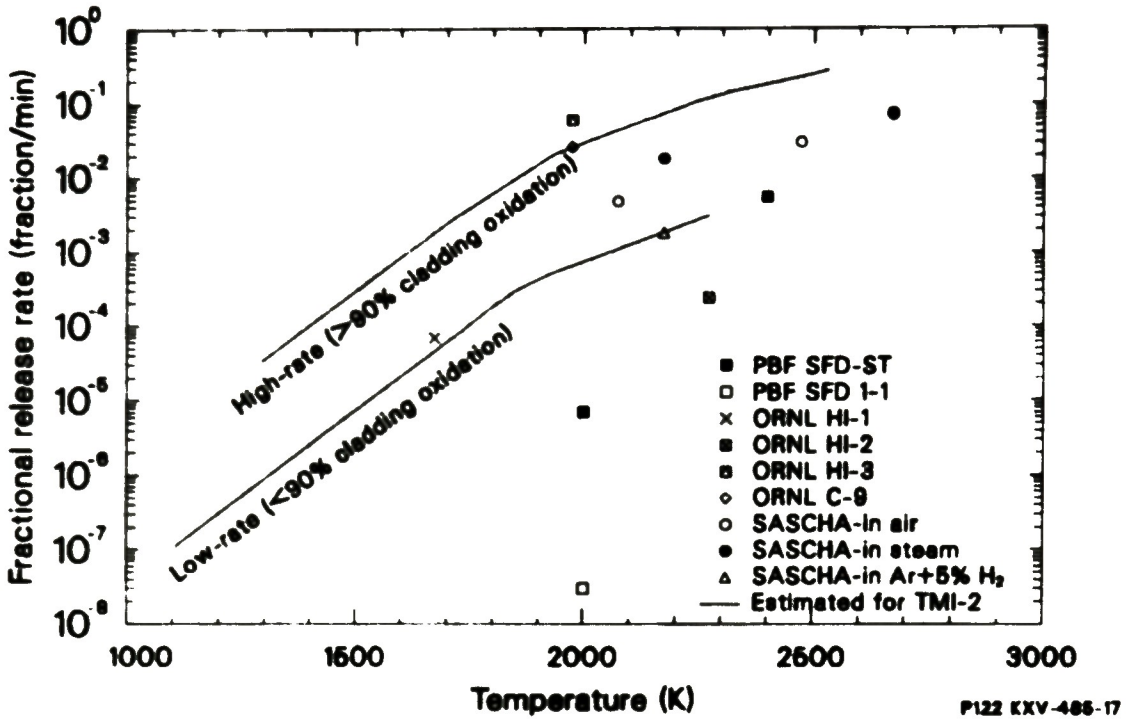

Figure 23. Comparison of fractional release rates.

TABLE 14. COMPARISON OF TMI-2 Te RELEASE FRACTIONS WITH IN-PILE AND OUT-OF-PILE TESTS

Event	Maximum Fuel Temperature (K)	Cladding Oxidation (%)	Release Fraction	Reference
TMI-2 Accident	2600	Low	5.8×10^{-2} (measured) 10.0×10^{-2} (estimated)	Present Study
PBF Tests				
SFD-ST	2400	100	4.0×10^{-1}	
SFD 1-1	2400	30	9.0×10^{-3}	
SFD 1-3	2400	30	1.4×10^{-4}	
ORNL Tests				
HI-1	1673	40	3.0×10^{-3}	2
HI-2	1973	100	0.5 to 1.0	2
HI-3	2273	35	6.0×10^{-3}	2
C-9	1973	100	9.7×10^{-1}	33
SASCHA Tests				
Low Steam Flow (1.5 L/min)	2573	Low	3.3×10^{-1}	2
High Steam Flow (30 L/min)	2733	High	6.5×10^{-1}	
Ar + 5% H_2	2173	0	3.6×10^{-2}	
Ar + 5% Steam	2200	Low	2.0×10^{-1}	

accident may have caused the holdup of the tellurium and ultimately low release fractions. Analyses of samples from the core region indicated a large fraction of tellurium was retained in the core.

CONCLUSIONS

Thermodynamic calculations were made, a number of available TMI-2 samples were analyzed, best estimate calculations were performed, and the data were compared with in-pile and out-of-pile tests. The following conclusions are drawn from the analysis:

1. Thermodynamic calculations indicate that H_2Te is the predominant vapor species in the upper plenum during the accident, however, at temperatures $\geq$1200 K, H_2Te dissociates to elemental tellurium.

2. Very little ($\sim$5.8%) tellurium was released and transported from the TMI-2 core, probably as a result of holdup by zircaloy cladding and structural materials. Analyses of samples from the core region indicated that a large fraction of tellurium was retained there.

3. Best estimate calculations suggest that a significant fraction of the total tellurium released was deposited on the upper plenum surfaces due to the high pressure injection at about 200 min after the reactor scram, resulting in high steam flow.

4. Comparison of tellurium release fractions and fractional release rates from the TMI-2 accident, with in-pile and out-of-pile test results, suggests that zircaloy holds tellurium until the cladding is oxidized significantly.

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APPENDIX A
TELLURIUM ANALYTICAL METHODS

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TELLURIUM ANALYTICAL METHODS

INTRODUCTION

The Idaho National Engineering Laboratory (INEL), as part of the broad core activities research program, will be conducting chemical analyses on a variety of samples obtained from the damaged Three Mile Island (TMI) Unit-2 reactor. The 1979 accident at TMI-2 resulted in the release of significant quantities of fission products from the damaged core to the reactor coolant system and containment building. Radioactive tellurium, as a radioiodine precursor, can constitute a significant health hazard if released to the environment. Also, the chemical interaction of tellurium with other reactor materials, principally zircaloy and stainless steel during severe core damage accidents, is not well understood. To date, there are very limited data available for tellurium in TMI-2 samples. Also, radioactive tellurium present in the fuel (except ^{125m}Te) at the time of the accident has now decayed to less than detectable concentrations. For this reason, attention has been focused on the postaccident measurement of stable fission product tellurium concentrations on reactor plant surfaces and core debris to determine tellurium retention and transport behavior.

Interpretation of quantitative tellurium data in TMI-2 samples is complicated by the fact that natural tellurium is routinely added to stainless steels as a free-machining agent.[1] The precise quantity of doped tellurium in a given sample of alloy is generally proprietary information; however, tellurium weight percentages of 0.0005 to 0.1% (5.0 to 1000 ppm) are typical. This interference requires that elemental analyses on all TMI-2 samples be screened for relative ratios of stainless steel components and tellurium (Fe/Ni/Cr/Te). In those cases in which tellurium an Fe/Ni/Cr are present in ratios which indicate the presence of doped steels, it will probably not be possible to extract any information relating to fission product tellurium. However, for those samples in which stainless steel components are not present in significant quantities, and for samples in which tellurium concentrations far exceed doped levels, tellurium analytical data may provide information on the behavior of fission product tellurium.

This Appendix provides a summary of analytical methods and procedures for the separation and analysis of tellurium in TMI-2 samples from the following areas:

- Leadscrews
- Makeup filter debris
- Reactor building basement debris and liquids
- Core debris (fuel, cladding, and structural materials).

The major elemental constituents of these core region materials include: iron, nickel, and chromium (304 and 17-4 PH stainless steel, Inconel 718 and x-750, zircaloy-4); zirconium (zircaloy-4); silver, cadmium and indium (absorber alloy); aluminum and boron (burnable poison rod); and uranium (fuel).

In some cases, a determination of tellurium concentration may be performed directly on a dissolved sample, without any preliminary treatment. However, this requires that: (a) the sample be adequate in size, with a tellurium concentration above instrumental detection limits: (b) no significant elemental interferences are present; and (c) the exposure rate of the sample is low enough to allow its use with the present radioactive exhaust system (currently 200 mR/h). For those samples where elemental interferences are present, or when preconcentration of the tellurium is necessary, the tellurium must be isolated from the bulk of the sample matrix.

EXPERIMENTAL METHODS

Analytical methods have been developed for the separation an analysis of tellurium in various sample matrices. Tellurium separation procedures have been evaluated, with ion-exchange resins, reductions, and precipitations providing the most effective means of separating tellurium

from complex matrices. Inductively Coupled Plasma-Atomic Emission Spectroscopy (ICP-AES) is the method of choice for the determination of total tellurium concentration.

Dissolution Methods

The TMI-2 samples from the core region have been brought into solution at the INEL through either a bisulfate fusion or a HNO_3/HF dissolution. The dissolution procedures for specific samples are described in detail in an analytical procedures report,[2] but a brief description of the dissolution procedures is reported here.

Bisulfate Fusion

Decomposition of a sample by the use of a fluxes is a common method for breaking up and/or oxidizing the components of a sample. A potassium bisulfate fusion has been successfully applied for the dissolution of a number of TMI-2 samples. The sample is slowly heated in a fusion chamber containing $Sr(NO_3)_2$ and $KHSO_4$, until the material fuses. After cooling, a known volume of deionized water is added to the melt. The insoluble $SrSO_4$ is removed by centrifuging and decanting the supernatant. An aliquot of the aqueous solution is available for ICP-AES analysis of tellurium.

HNO_3/HF Dissolution

Dissolution of metals and alloys containing tellurium presents no serious difficulties when carried out in HNO_3/HF solution at moderate temperatures (<100°C). A 1:1:1 volume mixture of water, nitric acid, and hydrofluroic acid is quite effective in the dissolution of high temperature alloys. Volatilization of tellurium is only a problem when a sample is warmed in the presence of a halogen, HCl, or HBr vapors.

In some cases, the HNO_3/HF dissolution procedure was used to leach and solubilize surface deposits from a base alloy sample. The dissolution was not allowed to proceed to the point where the base metal was

significantly attacked. However, the HNO_3/HF solutions typically can be expected to contain varying amounts of base metals along with the surface adherents. In these cases, the overall mass of the solubilized sample is not known. Analytical data on these samples will not provide absolute elemental concentrations (such as ag Te/g sample), but rather relative elemental constituents (such as wt%, normalized to a given major elemental component.

Analytical Method

A number of analytical methods are available for the determination of tellurium in a variety of inorganic materials. However, the convenience, sensitivity, and interferences of any given technique must be evaluated with consideration given to the samples of interest. The analyses method for the TMI-2 samples should: (a) allow for rapid qualitative elemental analyses (to screen for stainless steel content); (b) allow for rapid multielement quantitative analysis (to determine Fe/Ni/Cr/Te ratios); and (c) exhibit maximum sensitivity (to limit preconcentration in small samples). Gravimetric methods are inappropriate for the analysis of TMI-2 samples for several reasons, including limited accuracy and serious interference from heavy metals. Volumetric methods for tellurium determinations generally are based on direct or indirect redox titrations. However, they are most applicable for tellurium concentrations in the milligram range. Electrochemical methods for the determination of tellurium can be quite sensitive, but they require rigorous control of elemental oxidation states; lead and other metals pose serious interferences. X-ray fluorescence is capable of estimating tellurium concentration in alloys down to the 1-ppm range. However, the presence of selenium in the sample can dramatically affect the detection limit. Atomic adsorption spectroscopy is a convenient method for the analysis of small amounts of tellurium, but convenience is sacrificed for samples requiring multielement analysis.

Atomic emission spectroscopy utilizing an inductively coupled plasma excitation source (ICP-AES) is a particularly useful method for the rapid qualitative or quantitative analyses of multiple elements in a variety of

sample matrices. The ICP-AES instrument utilizes an inductively coupled argon plasma as an excitation source for the sample. Because of the high temperatures and high excitation power of plasmas, plasma emission spectra are extremely line rich. Qualitative analysis is accomplished by an analysis of the wavelengths of the emitted light; quantitative analysis relates the intensity of emitted light to the concentration of a given element. The ICP-AES instrument to be used for analyses of TMI-2 samples is a Leeman Labs Plasma-Spec.[3] This instrument has the following capabilities which make it appropriate for use with TMI-2 samples:

- Qualitative Analysis Mode--The use of this mode allows for rapid screening of a sample for up to 20 user-selected elements. This will allow an initial determination of sample matrix (dissolved stainless, zircaloy, control rods, etc.) in order to identify necessary background corrections.

- Quantitative Multielement Mode--The use of this mode allows for the rapid, sequential scanning of up to 20 wavelengths for user-selected elements. Concentrations of individual elements are calculated based on previously entered calibration curves. For most elements, the signal intensity exhibits excellent linearity over 3 to 5 orders of magnitude. This mode will allow for rapid quantitative analysis of all the elements of interest (Fe, Ni, Cr, Te, Zr, U, Cu, Sn, and Mo).

- Detection Limits--ICP-AES detection limits for elements to be analyzed in TMI-2 samples are presented in Table A-1. The detection limit for tellurium (15 ng/mL, 15 ppm) is the most critical, since trace quantities may be present in some samples. The concentration of natural tellurium doped in stainless steels (typically 5 to 1000 ppm) is well within the instrumental detection limit.

A number of the TMI-2 grab samples available for Te analysis are in the 10 to 200-mg size range. The dissolution procedure brought the total volume of these sample solutions to 20 mL. In order for the tellurium in

TABLE A-1. ICP-AES DETECTION LIMITS

Element	Detection Limit (ng/ml)
Tellurium (Te)	15.0
Iron (Fe)	0.09
Nickel (Ni)	0.2
Chromium (Cr)	0.08
Zirconium (Zr)	0.06
Uranium (U)	1.5
Tin (Sn)	3.0
Copper (Cu)	0.04
Molybdenum (Mo)	0.2

solution to be detectable without preconcentration, a minimum of 300 ng Te must be present. For the 10 to 200 mg samples, this requires approximately 1.5 to 30 ppm tellurium. Those samples in which the tellurium concentration is below the detection limit (as evidenced by the qualitative scans) will be retained for subsequent tellurium separation/concentration.

Separation Methods

The direct determination of TMI-2 samples may be prohibited by one or more of the following factors. First, the sample size and/or analytic concentration may be too small to reach the Te detection limit. Individual samples from the same bulk fraction may be consolidated if sample size is the limitation. A series of separations designed to concentrate the tellurium might be sufficient to alleviate the problem of analytic concentration. A second factor which might preclude the direct determination of tellurium would be the presence of significant quantities of interfering elements. Despite the fact that tellurium has 764 emission lines, only a limited number are appropriate for analytical use. High concentrations of elements with strong emission lines in the immediate vicinity of Te emissions might require that the tellurium be isolated from the interfering species prior to analysis. The final factor which may limit the direct determination of tellurium in TMI-2 samples is the effective radiation dose rate. A typical sample volume for multielement qualitative and quantitative analysis is approximately 5 to 10 mL. The majority of the total sample volume is released to a drain storage, and the remaining sample in the torch compartment is vented through a chimney to the radioactive exhaust system. The effective dose rate for an individual sample is limited to 200 mR/h. This may require that the tellurium be separated from some of the radioactive sample constituents, prior to analyses.

The most effective procedures for the separation and/or concentration of tellurium in TMI-2 samples are based on precipitations, reductions, and ion-exchange techniques. The most appropriate separation or concentration methods for a given sample can be selected based on the dose rate of the sample and the results of any preliminary qualitative scans.

Ion-Exchange

A method which makes use of a Dowex anion exchange resin is particularly useful for the separation of many of the species which may be present in TMI-2 samples.[4] The separation is carried out using a column of Dowex I x 8, 200 to 400 mesh, chloride form resin. The sample is taken up in an HF/HNO_3 solution (1:2 by volume) with a total fluoride concentration of 0.6 to 1.0 M. The pH of the resulting solution is adjusted to between 5 and 6 with a dilute solution of NH_4OH. Following a neutral wash of the column, the sample solution is introduced. A flow rate of 0.5 mL/min/cm^2 allows for adequate separation of individual species. A diagram of the separation scheme for individual elements is shown in Figure A-1, and efficiencies for selected elements are given in Table A-2.

Sn (II) Reduction

Microgram quantities of tellurium in aqueous acidic solution are very effectively reduced to the zero valient state by freshly prepared Sn (II) solutions.[5,6] This is widely used as a preconcentration method for ultimate tellurium determinations.

In order to minimize air oxidation of Sn (II), the following solution must be prepared daily. 100 mL of concentrated hydrochloric acid is added to 250 g of stannous chloride ($SnCl_2 \cdot 2H_2O$), and the solution is warmed until clear. When cool, the solution is diluted to 250 mL with concentrated hydrochloric acid.

Approximately 10 mL of the freshly prepared $SnCl_2$ solution is added to the warm acidic sample solution containing tellurium. The resulting solution is mixed, cooled to room temperature, and allowed to stand undisturbed for 3 to 4 h. The precipitated tellurium is filtered on a 0.6 μ micropore filter. The precipitation yield is 90% for samples containing <140 μg of tellurium.

A potential interference in the Sn (II) reduction method is the simultaneous reduction of As, Bi, Cu, Au, Hg, Se, and Ag to their metallic

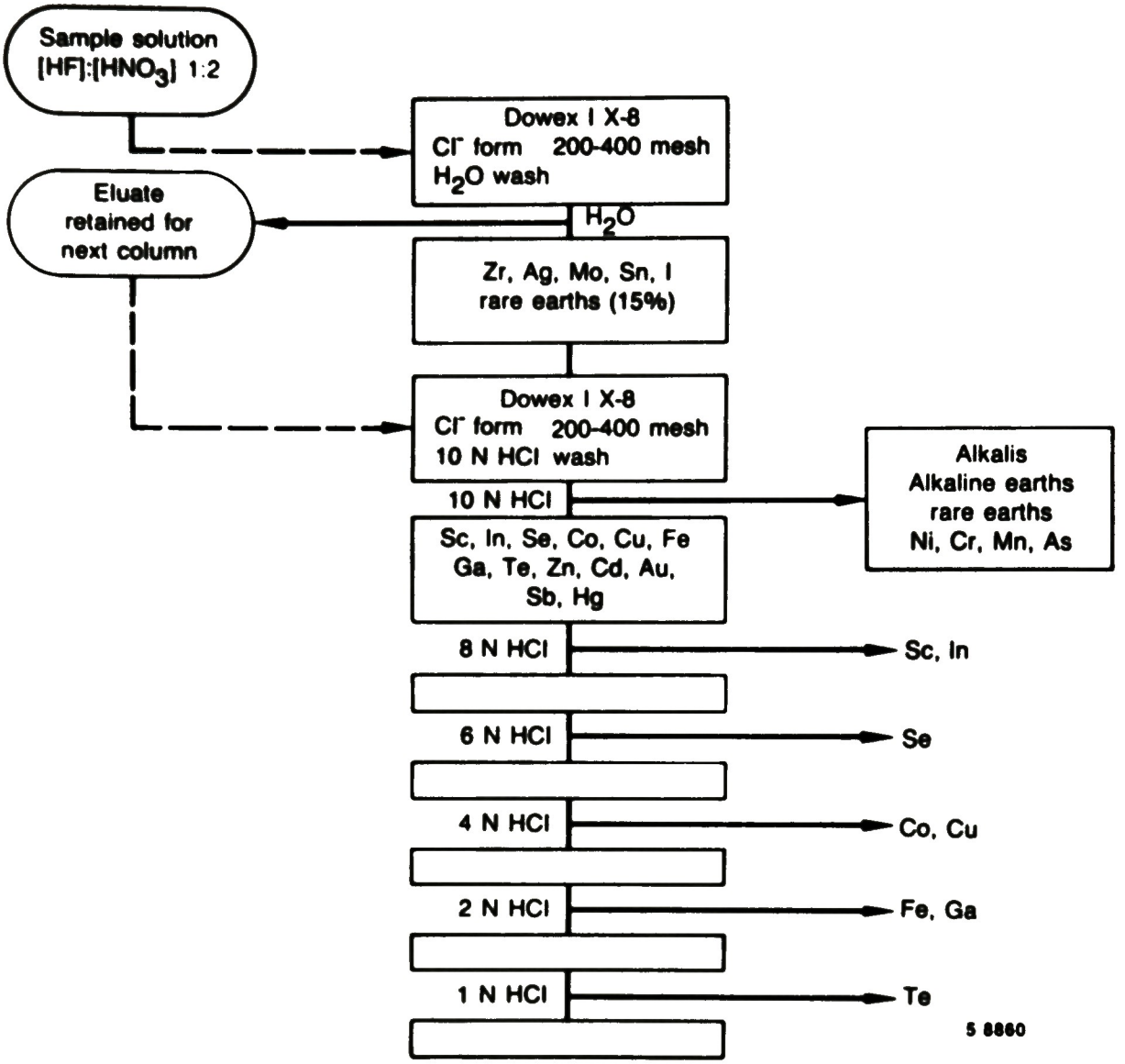

Figure A-1. Dowex separation scheme.

TABLE A-2. SEPARATION DATA USING DOWEX I X 8

Eluting Solvent	(Volume)[a]	Elute	Efficiency (%)[b]
8 N HCl	(12)	In	100
6 N HCl	(5)	Se	100
4 N HCl	(6)	Co,Cu	100
2 N HCl	(8)	Fe	95
1 N HCl	(5)	Te	100

a. The number in parentheses represents the number of column volumes of eluting solvent necessary to effect the separation.

b. The efficiency is reported as the percentage of the total element eluted by the solvent at the moment of elution ($\pm$0.1%).

states. This technique is not generally appropriate for the separation of analytically pure tellurium, but is very useful for the preconcentration of microgram quantities.

SO_2 Reduction

The reduction of tellurium by SO_2 in acidic solution is the preferred method for obtaining analytically pure tellurium metal.[7] Although the reduction can be carried out with SO_2 alone, this requires that no HNO_3 be present in solution, and all the tellurium be in the +4 oxidation state. The limitation can be avoided by the use of SO_2 and hydrazine, which effectively reduces either the +4 tellurite or the +6 tellurite to the zero valient metal.[8]

Fifteen mL of a saturated SO_2 solution is added to a warm HCl solution (2.0 to 5 N) containing tellurium. The reducing agent can be added by bubbling SO_2 through the solution, however this results in the loss of significant quantities of $TeCl_2$ through volatilization. Approximately 10 mL of 15% $N_2H_4 \cdot HCl$ is added to the SO_2/Te solution, followed by another 25 mL of saturated SO_2 solution. The resulting solution is brought to a boil, and allowed to boil until the Te precipitate settles out of solution (typically less than 5 min). The slow precipitation of tellurium from a warm solution allows for the formation of a more crystalline precipitate, which is more resistant to air oxidation.

This reduction procedure results in the quantitative precipitation of elemental tellurium if the solution HCl concentration is 2.0 to 5.0 N.[9] The tellurium does not precipitate from >8 N HCl solutions. Mercury may be coprecipitated along with the tellurium.

Fe (III) Coprecipitation

Coprecipitation of tellurium with metal hydroxides is a very effective means of concentrating small amounts of tellurium in acidic solution.[10] The method is equally effective for Te (IV) and Te (VI), and is best

accomplished in solutions containing less than 400 mg of iron. Tellurium can be precipitated from HNO_3 by this method, but the solution must not contain more than 3% (by volume) HNO_3.

The slightly acidic sample solution containing 100 to -200 mg of $Fe(NO_3)_3$ is poured into an excess of NH_4OH solution (pH 9.7) with constant stirring. As the $Fe(OH)_3$ precipitates, up to 0.5 mg of tellurium is quantitatively precipitated. The precipitation yield lowers to 65-80% for samples containing less than 0.1 mg of tellurium. Also, the coprecipitation efficiency of tellurium is dramatically affected by pH. For example, only a 65% efficiency is attained at a pH of 8.0. Because of this, it is important that the acidic sample be added to an excess of base solution, and the pH of the mixture be monitored during the course of the precipitation.

The presence of Zn, Cd, and Mo in large amounts does not interfere with tellurium coprecipitation, however, more than 400 mg of iron causes difficulty in the filtration of the hydroxide precipitate. Lead may be coprecipitated along with the tellurium.

Metal Sulfide Precipitation

The precipitation of tellurium sulfide affords a convenient means of separating tellurium from those elements which form soluble sulfides, including Fe, Ni, Cs, and Sr. The major interference in the method is presented by copper, so an initial qualitative test for its presence must be run. If necessary, copper can be removed by precipitation with hydroquinone.

Te (IV) is very readily reduced by H_2S, but Te (VI) is considerably slower. In order to convert all the Te (VI) in acid solution to the +4 oxidation state, the solution is taken up in dilute (<6 N) HCl and warmed (at <100°C). This acidic solution of Te (IV) is then saturated with H_2S by slowly bubbling the gas through the solution. It is important that the TeS_2 precipitation be done cold, in order that the resulting precipitates are reasonably soluble.

Discussion

The tellurium analytical methods described in this appendix have been selected for their applicability toward tellurium determinations in a series of samples from the damaged TMI Unit-2 reactor. The materials which will be analyzed for tellurium content include stainless steels, inconels, zircaloy-4, control rod alloy, burnable poison, fuel, and coolant. These matrices are found in samples from leadscrews, makeup filter debris, reactor building basement debris and liquids, and core debris (fuel, cladding, and structural materials).

The direct application of tellurium analytical data toward an explanation of fission product deposition is complicated by the fact that there may be serious contamination of TMI-2 samples by natural tellurium. Tellurium is routinely present in stainless steels in weight percentages of 0.00050 to ~0.1%. As a consequence, all elemental analyses for TMI-2 samples must be screened for relative amounts of stainless steel components. Analytical tellurium data from samples which do not contain significant quantities of stainless steel components (Fe/Ni/Cr) may be interpreted as arising from fission product tellurium with a reasonable degree of certainty. Those samples which contain appreciable quantities of stainless steel components must be carefully evaluated to determine the significance of any tellurium data in terms of fission product behavior. If the tellurium and Fe/Ni/Cr are present in ratios which approximate those found in doped steels, it will not be possible to extract any information about fission product tellurium. However, if the tellurium concentration far exceeds the 0.1 wt% maximum for doped steels, some qualitative information about fission product tellurium can be inferred. In this case quantitative fission product tellurium information is not possible since the concentrations of doped tellurium can vary over three orders of magnitude.

The analytical method to be used for the determination of tellurium in TMI-2 samples is Inductively Coupled Plasma-Atomic Emission Spectroscopy (ICP-AES). The instrumental detection limits for the elements of interest (Table A-1) are within the ranges necessary for the analysis of most of the

TMI-2 samples. In some cases, when tellurium concentrations are below instrumental detection limits, a series of separations can be used which are designed to preconcentrate the tellurium.

ICP-AES interferences generally consist of spectral overlap or matrix effects. In most cases, serious interferences arising from spectral overlap can be avoided by using alternate analytical lines. For example, the determination of iron in the presence of uranium requires that a somewhat unusual iron line be selected in order to avoid spectral interferences from uranium at other wavelengths.[11] It is important that potential spectral interferences be evaluated in any given sample based on the known elemental composition, or a qualitative survey of individual constituents.

In general, ICP-AES systems are comparatively free from matrix effect, at least when compared to atomic absorption spectroscopy. In most cases, matrix matching samples with standards will reduce the interferences to acceptable levels. For the TMI-2 samples a series of high purity multielement ICP standards containing Fe, Ni, Cr, and Te have been prepared, as well as standards containing Fe, Ni, Cr, and Te in acid solutions of UO_2 and zirconium. In this way, standards will be matrix matched with the major elemental constituents present in core region materials. The standard calibration curves will be selected for a matrix based on the qualitative elemental scan. This should essentially eliminate any matrix interferences in the TMI-2 samples.

The individual separation procedures described in this appendix are applicable toward a variety of analysis problems, however, the most effective selection of appropriate methods for an individual sample can be based on specific factors. A summary of the individual analysis steps would include the following:

1. Determination of effective radioactive dose rate for a sample solution.

   Sample solutions for ICP-AES analysis (typically about 5 mL) are limited to a 200 mR/h dose rate. Samples which exceed this value

will require some preliminary separations to isolate the tellurium from the bulk of the sample activity. The Dowex ion-exchange method, Sn (II) reduction, or iron hydroxide coprecipitation might be appropriate for such an initial separation.

2. Perform a rapid qualitative elemental scan for elements of interest (Te, Fe, Ni, Cr, Zr, Cu, Sn, Mo).

A rapid qualitative scan of the sample serves several purposes. First, it provides a quick evaluation of whether the tellurium concentration is above or below the instrumental detection limit. The absence of a tellurium signal indicates that preconcentration of the solution may be necessary. Alternatively, in the case of very small samples, it may be possible to consolidate several small samples from the same bulk fraction.

Another purpose of a qualitative sample analysis is the evaluation of individual sample matrices. The identification of the major elemental constituents in a sample serves to target the origin of the material [zircaloy is indicated by tin, 17-5 PH (stainless steel) is indicated by copper, inconel is indicated by molybdenum], and provide the sample matrix for the evaluation of elemental interferences in subsequent procedures.

Based on the results of the qualitative analysis, a sample is either targeted for preconcentration/separation, or available for quantitative analysis.

3. Perform a sequential multielement quantitative analysis for elements of interest.

Quantitative data for tellurium, iron, nickel, and chromium should be obtained for each sample in which the tellurium concentration is above the instrumental detection limit. Samples

which contain significant quantities of fission product tellurium (as determined by the Te scan and Fe/Ni/Cr ratios) may be more comprehensively analyzed by performing quantitative analyses for those elements which comprise the sample matrix.

Experimental molar ratios of selected elements for individual samples can be compared with nominal ratios in core materials to determine the origin of a given sample matrix. A summary of elemental composition for selected core materials is tabulated in Table A-3.

4. Perform tellurium separation or preconcentration procedures as necessary.

Separation of tellurium from individual elements may be necessary for two reasons. First, the dose rate of the sample may be too high (>200 mR/h) to allow its use in the ICP-AES system. The major contributing isotopes to the sample activity can be identified, and separation schemes selected which will effectively isolate the tellurium from the bulk of the sample activity. In most cases, separation of the Cs and Sr would probably be sufficient to lower the dose rate to acceptable levels.

Separation or preconcentration of tellurium from the bulk of the sample matrix may also be necessary when its concentration is below the instrumental detection limit. The experimental procedures described in this appendix are applicable toward many sample matrices. However, selection of the individual method is most effectively based on the results of a qualitative elemental scan. For example, the Fe (III) coprecipitation method is only effective for samples containing less the 400 mg of iron. This probably precludes use of this technique for samples containing large amounts of steels. Also, the ion-exchange method is limited in its application to large samples by the loading capacity of the resin.

TABLE A-3. MATERIAL COMPOSITION OF TMI-2 CORE REGION COMPONENTS (wt%)

Material	Fe	Ni	Cr	Cu	Sn	Mo
304 SS	Major	8-10.5	18-20	--	--	--
17-4 PH	Major	3-5	15.5-17.5	3-5	--	--
Zircaloy-4	0.2	--	0.1	--	1.5	3
	Ag	Cd	In	Al_2O_3	B_4C	
Control Rod	80	5	15	--	--	
Burnable Poison	--	--	--	98	1.5	

Because of the relatively high efficiencies of the separation techniques (65 to ~100% depending on the experimental conditions), these methods should serve to isolate tellurium in quantities necessary for ICP-AES analysis.

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APPENDIX B

TRAP-MELT INPUT DECK FOR TMI-2

APPENDIX B
TRAP-MELT INPUT DECK FOR TMI-2

This appendix documents the thermal hydraulic data, control volume geometries, and tellurium release rates used to setup the TRAP-MELT input deck for TMI-2. The start and total times were from 9180 and 12780 s. Input cards 23, 24, 25, 26, 27, and 30 in the input deck are for control volume geometric, thermal hydraulic and source data, respectively.

INPUT DECK FOR TMI-2 CONTROL VOLUMES CONSISTS OF:

```
PKBKV,P1,T7777,STHLC,EC124.
ACCNT,ID=PK8,CHG=756101030,BIN=TB5.
COMMENT.
COMMENT.
COMMENT.   K. V. JOB
COMMENT.   PF=PKBKV11,ID=PKBKV
COMMENT.
COMMENT.   THIS IS RUN #12 FOR 3600 SEC
COMMENT.
COMMENT.
COMMENT.
COMMENT.
COMMENT.   GET THE TRAP-MELT CODE
COMMENT.
ATTACH,TRAP,TRAPTEMP,ID=DVT,CY=2,MR=1.
COMMENT.
COMMENT.   GET EXTRA MEMORY
COMMENT.
RFL,EC=124.
COMMENT.
COMMENT.   RUN THE PROGRAM
COMMENT.
TRAP.
EXIT,U.
REDUCE,ECS.
REQUEST,NEW,*PF.
CATALOG,CWAFI,TRAPCWAFIDATAKVRUN11,ID=PKBKV,RP=30,MR=1.
BEGIN,MASTER.
REWIND,CWAFO.
CATALOG,CWAFO,TRAPCWAFODATAKVRUN11,ID=PKBKV,RP=30,MR=1.
COMMENT.
COMMENT.    COPY OUTPUT TO PERM-FILE FOR POSSIBLE REPRINTING LATER
COMMENT.
EXIT,U.
REWIND,INPUT.
COPYSBF,INPUT,OUTPUT.
REWIND,OUTPUT.
COPY,OUTPUT,NEW.
REWIND,NEW.
CATALOG,NEW,TRAPOUTPUTLISTKVRUN11,ID=PKBKV,RP=30,MR=1.
RETURN,NEW.
*
*
*---------------------------------------------------------------
*
*
*
*RHR PIPE DIVIDED INTO ONE CONTROL VOLUMES
*
*LOWER PORTION OF UPPER PLENUM CONTROL
*       VOLUME INCLUDES CENTRAL ASSEMBLY ONLY
*
*CP TIME BEFORE WRITING RESTART FILE
*CARD C
*CPMAX BELOW
4076
*NUMBER OF PRINT INTERVALS
*CARD 1
12
*PRINT COMMENT CARDS? (YES=1,NO=0)
*CARD 2
0
*PRINT EVAPORATION RATES? (YES=1,NO=0)
*CARD 3
0
*RESTART FLAG(RESTARTED JOB=1,NEW JOB=0)
*CARD 4
0
*START TIME,TOTAL PROBLEM TIME,MAX TIMESTEP (SECONDS)
*CARD 5
9180.0 12780.0 1.0
*CONVERGENCE PARAMETERS(REL,ETA1,ETA2)
*CARD 6
.0001 .001 0.1
*NO. SPECIES(USE 5),NO. CONTROL VOLUMES,NO. STATES(=5)
*    NO. PARTICLE DISTRIBUTION PARAMETERS(=2)
*CARD 7
5 8 5 2
*SPECIES IDS(NO. IDS SHOULD EQUAL NO. OF SPECIES)
*CARD 8
I2 CI CH PU TE
*
*FLOW CONNECTIONS
```

```
*
*CONTROL VOLUME -  NUMBER
*
* LOWER PLENUM          1
* CORE                  2
* UPPER PLENUM          3
* UPPER HEAD            4
* HOT LEG               5
* PRESSURIZER           6
* STEAM GENERATOR       7
* COLD LEG              8
*
*CARD 9S
*
CCOOOOC1
100CC0C0
C1000000
00100000
00100CC0
C00C10C0
0000 10C0
C00CC01C
*
*NO. PROGRAM FLAGS IN BETV(*3)
*CARD 10
3
*VAPOR SORPTION,PARTICLE SETTLING FLAGS
*3 COLUMNS 1 LINE FOR EACH CONTROL VOLUME
*COLUMN 1 - VAPOR SORPTION FLAG(YES=1,NO=0)
*COLUMN 2 - PARTICLE DEPOSITION FLAG(YES=1,NO=0)
*COLUMN 3 - FLOW/PARTICLE SETTLING DIRECTION FLAG
*            1=CROSS FLOW SETTLING (WITH THE FLOW)
*            0=COUNTERFLOW SETTLING(AGAINST)
*CARD 11S
110
CC0
100
100
110
100
100
110
*NO. CONTROL FLAGS IN ADHOC(*1)
*CARD 12
1
*CONTROL FLAG FOR VAPOR CONDENSATION(YES=1,NO=0)
*     1 LINE FOR EACH CONTROL VOLUME
*CARD 13S
1
0
1
1
1
1
1
1
*COAGULATION CONTROL *ORDING, DIGITS=NO. CONTROL VOLUMES)
*     A 1 IN THE N-TH DIGIT PERMITS COAGULATION IN
*     THE N-TH CONTROL VOLUME
*CARD 14
00111100
*ARBITRARY MULTIPLIERS(SET=1.0)
*CARDS 15-21
1.0
1.0
1.0
1.C
1.0
1.0
1.0
*TURBULENT COAGULATION FLAG(YES=1,NO=0)
*CARD 22
1
*CONTROL VOLUME GEOMETRIC PARAMETERS
*
*COMPILED BY K.VINJAMURI
*
*CONTROL VOLUMES 1-8
*LENGTH-HYDRAULIC DIAMETER-FLOW AREA-SETTLING AREA-HEIGHT
*CARD 23S
  0.07      C.73       159.2      184.7          8.07
 12.C0      0.042       52.3        1.0E-1C      12.0
 11.52      0.43        34.8       90.0          11.52
  5.61      C.43        34.0      124.0           5.61
 82.00      0.023        7.1       42.4          82.0
 45.00      0.16        38.5        1.0E-10      45.0
104.30      C.C46       52.4        1.0E-10     104.3
 52.00      C.011        4.26      23.8          52.0
```

```
*-----------------------------------------------------
*FLOW RATE DATA BETWEEN CONTROL VOLUMES AT FLOW
*      JUNCTIONS--FLOW JUNCTIONS ARE NUMBERED 1
*      THRU 7--JUNCTION 1 INDICATES FLOW BETWEEN
*      CONTROL VOLUME 1 AND 2,JUNCTION 2 BETWEEN
*      CONTROL VOLUMES 2 AND 3,ETC.
*
*FLOW IS IN LBM/SEC
*
*CARD 24S
*CARD 24S INCLUDE THREE DATA SETS FOR EACH JUNCTION
*      CARD 24A INDICATES NUMBER OF DATA ENTRIES
*      CARD 24B CONTAINS THE TIME DATA POINTS
*      CARD 24C CONTAINS THE MASS FLOW RATES
*JUNCTION 1 (VOL.8 TO VOL.1)
13
9180.0 9480.0 9780.0 10080.0 10380.0 10680.0
10980.0 11280.0 11580.0 11880.0 12180.0 12480.0 12780.0
1.0 1.0 1.0 1.0 3.3E+03 3.3E+03
1.0 1.0 1.0 1.0 1.0 1.0 1.0
*
*JUNCTION 2 (VOL.1 TO VOL.2)
13
9180.0 9480.0 9780.0 10080.0 10380.0 10680.0
10980.0 11280.0 11580.0 11880.0 12180.0 12480.0 12780.0
1.0 1.0 1.0 1.0 3.3E+3 3.3E+3
1.0 1.0 1.0 1.0 1.0 1.0 1.0
*
*JUNCTION 3 (VOL.2 TO VOL.3)
13
9180.0 9480.0 9780.0 10080.0 10380.0 10680.0
10980.0 11280.0 11580.0 11880.0 12180.0 12480.0 12780.0
20.0 15.12 12.57 8.62 3356.0 3356.0
45.06 5.09 2.55 1.84 78.3 78.3 78.3
*
*JUNCTION 4 (VOL.3 TO VOL.4)
13
9180.0 9480.0 9780.0 10080.0 10380.0 10680.0
10980.0 11280.0 11580.0 11880.0 12180.0 12480.0 12780.0
20.0 15.12 12.57 8.62 3356.0 3356.0
45.06 5.09 2.55 1.84 78.3 78.3 78.3
*
*JUNCTION 5 (VOL.3 TO VOL.5)
13
9180.0 9480.0 9780.0 10080.0 10380.0 10680.0
10980.0 11280.0 11580.0 11880.0 12180.0 12480.0 12780.0
1.0 1.0 1.0 1.0 3333.0 3333.0
1.0 1.0 1.0 1.0 1.0 1.0 1.0
*
*JUNCTION 6 (VOL.5 TO VOL.6)
13
9180.0 9480.0 9780.0 10080.0 10380.0 10680.0
10980.0 11280.0 11580.0 11880.0 12180.0 12480.0 12780.0
1.0 1.0 1.0 1.0 3333.0 3333.0
1.0 1.0 1.0 1.0 1.0 1.0 1.0
*
*JUNCTION 7 (VOL.5 TO VOL.7)
13
9180.0 9480.0 9780.0 10080.0 10380.0 10680.0
10980.0 11280.0 11580.0 11880.0 12180.0 12480.0 12780.0
1.0 1.0 1.0 1.0 3333.0 3333.0
1.0 1.0 1.0 1.0 1.0 1.0 1.0
*JUNCTION 8 (VOL.7 TO VOL.8)
13
9180.0 9480.0 9780.0 10080.0 10380.0 10680.0
10980.0 11280.0 11580.0 11880.0 12180.0 12480.0 12780.0
1.0 1.0 1.0 1.0 3333.0 3333.0
1.0 1.0 1.0 1.0 1.0 1.0 1.0
*
*-----------------------------------------------------
*
*CARDS 25,26AND 27 CONTAIN GAS PRESSURE,GAS TEMPERATURE,
*      AND WALL TEMPERATURE DATA FOR EACH CONTROL VOLUME
*
*      FOR EACH CONTROL VOLUME A CARD 25 SET IS INPUT
*      THEN A CARD 26 SET THEN A CARD 27 SET
*      THIS SEQUENCE IS REPEATED FOR EACH CONTROL VOLUME
*
*CARD 25 SET CONSISTS OF:
*        CARD 25A INDICATING THE NUMBER OF PRESS DATA PTS.
*        CARD 25B CONTAINING THE TIME DATA POINTS
*        CARD 25C CONTAINING THE PRESSURE DATA ENTRIES(PSI)
*
*CARD 26 SET CONSISTS OF:
*        CARD 26A INDICATING THE NUMBER OF GAS TEMP ENTRIES
*        CARD 26B CONTAINING THE TIME DATA POINTS
*        CARD 26C CONTAINING THE GAS TEMP ENTRIES(DEG. F)
*
```

```
*CARD 27 SET CONSISTS OF:
*         CARD 27A INDICATING THE NUMBER OF WALL TEMP ENTRIES
*         CARD 27B CONTAINING THE TIME DATA POINTS
*         CARD 27C CONTAINING THE WALL TEMP DATA ENTRIES(DEG.F)
*
*CONTROL VOLUME 1 (LOWER PLENUM)
*
13
9180.0 9480.0 9780.0 10080.0 10380.0 10680.0
10980.0 11280.0 11580.0 11880.0 12180.0 12480.0 12780.0
825.4 921.5 1050.8 1196.2 1755.0 2066.2
2078.5 2126.6 1937.7 1914.4 1584.5 1472.3 1507.1
*
1
0.0
590.0
*
1
0.0
540.0
*
*CONTROL VOLUME 2(CORE)
*
13
9180.0 9480.0 9780.0 10080.0 10380.0 10680.0
10980.0 11280.0 11580.0 11880.0 12180.0 12480.0 12780.0
825.4 921.5 1050.8 1196.2 1755.0 2066.2
2078.5 2126.6 1937.7 1914.4 1584.5 1472.3 1507.1
*
1
0.0
630.0
*
1
0.0
540.0
*
*CONTROL VOLUME 3( UPPER PLENUM )
*
13
9180.0 9480.0 9780.0 10080.0 10380.0 10680.0
10980.0 11280.0 11580.0 11880.0 12180.0 12480.0 12780.0
825.4 921.5 1050.8 1196.2 1755.0 2066.2
2078.5 2126.6 1936.7 1914.4 1584.5 1472.3 1507.1
*
1
0.0
1100.0
*
1
0.0
1105.0
*
*CONTROL VOLUME 4(UPPER PLENUM HEAD )
*
13
9180.0 9480.0 9780.0 10080.0 10380.0 10680.0
10980.0 11280.0 11580.0 11880.0 12180.0 12480.0 12780.0
825.4 921.5 1050.8 1196.2 1755.0 2066.2
2078.5 2126.6 1936.7 1914.4 1584.5 1472.3 1507.1
*
1
0.0
800.0
*
1
0.0
805.0
*
*CONTROL VOLUME 5(HOT LEG)
*
13
9180.0 9480.0 9780.0 10080.0 10380.0 10680.0
10980.0 11280.0 11580.0 11880.0 12180.0 12480.0 12780.0
825.4 921.5 1050.8 1196.2 1755.0 2066.2
2078.5 2126.6 1936.7 1914.4 1584.5 1472.3 1507.1
*
1
0.0
630.0
*
1
0.0
630.0
*
*CONTROL VOLUME 6(PRESSURIZER)
13
9180.0 9480.0 9780.0 10080.0 10380.0 10680.0
```

```
10980.0 11280.0 11580.0 11880.0 12180.0 12480.0 12780.0
825.0 922.0 1051.0 1196.0 1750.0 2066.0
2079.0 2127.0 1937.0 1914.0 1585.0 1472.0 1507.0
*
1
0.0
630.0
*
*
1
0.0
635.0
*
* CONTROL VOLUME 7(STEAM GENERATOR)
*
13
9180.0 9480.0 9780.0 10080.0 10380.0 10680.0
10980.0 11280.0 11580.0 11880.0 12180.0 12480.0 12780.0
825.0 922.0 1051.0 1196.0 1750.0 2066.0
2079.0 2127.0 1937.0 1914.0 1585.0 1472.0 1507.0
*
1
0.0
630.0
*
1
0.0
635.0
*
*CONTROL VOLUME 6(COLD LEG)
*
13
9180.0 9480.0 9780.0 10080.0 10380.0 10680.0
10980.0 11280.0 11580.0 11880.0 12180.0 12480.0 12780.0
825.0 922.0 1051.0 1196.0 1750.0 2066.0
2079.0 2127.0 1937.0 1914.0 1585.0 1472.0 1507.0
*
1
0.0
600.0
*
1
0.0
610.0
*
*END OF PRESSURE/TEMPERATURE INPUT
*
*-------------------------------------------------------------
*
*
*BINARY CODED WORDS FOR INITIAL MASSES PRESENT AT START OF
*       PROBLEM(NUMBER OF DIITS EQUALS NUMBER OF CONTROL
*       VOLUMES/ONE WORD FOR EACH SPECIES)
*       FOR KV PROBLEM ALL ARE ZERO SINCE NO INITIAL MASSES
*       ARE PRESENT
*CARD 28S
*
00000000
00000000
00000000
00000000
00000000
*
*CARD 29 IS OMITTED SINCE ALL VALUES EQUAL ZERO ON CARD 28S
*
*
*
*SOURCE DATA INPUT
*
*CARD 30S
*
*THE FOLLOWING CARDS ARE REPEATED FOR EACH SPECIES
*    THAT HAS A SOURCE
*
*          CARD 30A-BINARY CODED WORD(NUMBER DIGITS EQUALS
*                   NUMBER OF CONTROL VOLUMES)
*                   A 1 IN THE N-TH DIGIT INDICATES A SOURCE
*                   EXISTS IN THE N-TH CONTROL VOLUME
*          CARD 30B-BINARY CODED WORD(NUMBER OF DIGITS EQUALS
*                   THE NUMBER OF STATES(=5)
*                   A 1 IN THE N-TH DIGIT INDICATES THAT A
*                   SOURCE EXISTS IN THE N-TH STATE
*                   NOTE: STATE 1=VAPOR IN GAS PHASE
*                         STATE 2=PARTICLE IN GAS PHASE
*                         STATE 3=VAPOR CONDENSED ON WALL
*                         STATE 4=PARTICE DEPOSITED ON WALL
*                         STATE 5=VAPOR REACTED ON WALL
*
```

```
*     (CARD 308 SET THE FOLLOWING CARDS ARE REQUIRED )
*
*          CARD 30C-NUMBER OF SOURCE RATE ENTRIES
*          CARD 30D-TIME DATA POINTS
*          CARD 30E-SOURCE RATE DATA ENTRIES(G/SEC)
*
*          30C,30D,30E ARE REPEATED FOR EACH STATE WITH A SOURCE
*          IN THE FIRST VOLUME,THEN THE SECOND VOLUME,ETC.
*
*ALL SOURCES ARE FROM
*
*I2 SOURCE INFORMATION IN (CONTROL VOLUME 2)
*
*AS A VAPOR IN THE GAS PHASE
*
*
C1000000
1C0C0
*
1
9180.0
.38849
*
*
*     * END OF I2 SOURCE INFORMATION *
*
*
*CESIUM IODIDE SOURCE INFORMATION
*
*AS A VAPOR IN THE GAS PHASE
*VERY SMALL CESIUM IODIDE SOURCE IS ASSUMED
*
01CC00C0
10000
*
1
9180.0
0.398
*
*
*     * END OF CESIUM IODIDE SOURCE INFORMATION *
*
*
*CESIUM HYDROXIDE SOURCE INFORMATION
*
*AS VAPOR IN GAS PHASE
*VERY SMALL CESIUM HYDROXIDE SOURCE INFORMATION
*
01000000
10000
*
1
9180.0
1.235
*
*
*     * END OF CESIUM HYDROXIDE SOURCE INFORMATION *
*
*
*SILVER-INDIUM-CADMIUM CONTROL MATERIAL AEROSOL SOURCE INFO.
*
*
*PARTICLES IN GAS PHASE ( CONTROL VOLUME 2)
*
C10CC0C0
010CC
*
1
9180.0
26.94
*
*
*   * END OF AEROSOL SOURCE INFORMATION *
*
*TELLURIUM SOURCE INFORMATION
*
*AS A VAPOR IN THE GAS PHASE  (CONTROL VOLUME 2)
*
*
01000000
100CC
*
```

```
1
9160.0
1.0119E-01
*
*
*END OF SOURCE DATA
*
*PARTICLE SOURCE SIZE DISTRIBUTION INPUT
*
*          PARTICLE SOURCES ARE ASSUMED LOG-NORMALLY DISTRIBUTED
*           WITH PARAMETERS(SIGMA-G/GEOMETRIC STANDARD DEVIATION,
*           RG/GEOMETRIC MEAN)
*
*CARD 31S
*
*            CARD 31A-BINARY CODED WORD WITH NUMBER OF DIGITS EQUAL
*                     THE NUMBER OF CONTROL VOLUMES
*                     A 1 IN THE N-TH DIGIT INDICATES THAT THE
*                     PARTICLE SOURCE DISTRIBUTION PARAMETERS
*                     ARE READ IN FOR THE N-TH VOLUME
*
*           NOTE: ALL PARTICE SOURCES(FROM ALL SPECIES) HAVE THE
*                  SAME DISTRIBUTION WITHIN A GIVEN CONTROL VOLUME
*
*         THE FOLLOWING CARD ARE REPEATED FOR EACH CONTROL VOLUME
*           INDICATED ON CARD 31A
*
*            CARD 31B-NUMBER OF SIGMA-G ENTRIES
*            CARD 31C-TIME DATA POINTS FOR SIGMA-G ENTRIES
*            CARD 31D-SIGMA-G ENTRIES
*
*            CARD 31E-NUMBER OF RG ENTRIES
*            CARD 31F-TIME DATA POINTS FOR RG ENTRIES
*            CARD 31G-RG ENTRIES(MICROMETERS)
*
01000000
*
*
*SIGMA-G TABLE
1
0.0
1.7
*
*RG TABLE
1
0.0
0.05
*
*AEROSOL DENSITY(G/CM**3) IN EACH VOLUME
*
*CARD 32
*
 2.5 2.5 2.5 2.5 2.5 2.5 2.5 2.5
DEMUX,COSCRA
MERGE
LFN,2,CHAFI
LFN,3,CHAFO
SU,2
SU,3
RETURN
RETURN


**********          PKB0003  //// END OF LIST ////
**********          PKB0003  //// END OF LIST ////
```